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Nonlinear Index of Refraction

Many of the interesting phenomena of nonlinear optics derive their behavior from the nonlinear index of refraction. It has led to a variety of fascinating applications. It is also central to many fundamental scientific investigations, such as chaos theory and quantum optics.

Several diverse physical effects contribute to the nonlinear index of refraction. Generally, it can be defined as a change in the refractive index or the spatial distribution of the refractive index of a medium due to the presence of optical waves. There are several different types of effects that fall under this general definition.

Optical Kerr effect. This effect is directly related to the third order nonlinear polarization. The change in index is proportional to the time average of the optical electric field squared, $\langle \mathbf{E}^{(\sim)}(\mathbf{r}, t) \cdot \mathbf{E}^{(\sim)}(\mathbf{r}, t) \rangle$. Another way to say this is that the change in index depends linearly on the optical intensity.

Nonlocal effects. In this case, the index change again depends on the time averaged field, or the intensity, but is due to a gradient in the field or long range order in the medium. The gradient could be caused by, for example, a focused beam or the superposition of coherent beams producing an interference pattern of bright and dark fringes. Long range order exists, for example, in liquid crystals. In these materials, if a molecule experiences a force due to the presence of an electromagnetic wave, intermolecular interactions cause molecules a relatively long distance away to be also affected by the force.

Saturation. In this type of effect, the change in index accompanies absorption of optical energy by the medium. Thus this case involves optical loss. For small enough intensities, the index change is approximately proportional to the intensity.

Changes in physical properties of a medium. In some instances an intense optical wave will induce changes in some physical property of a medium. Examples are the temperature or the concentration of a particular species. The index change will often be proportional to the intensity but may also depend on the time integrated intensity, that is, the fluence.

Cascading effects. An effective change in index can result from a combination of second order effects, or a sequential effect of linear and second order phenomena. An example of the former is the change in optical phase of the pump wave in a general second harmonic generation or frequency mixing process. The photorefractive effect, discussed below, is an example of the latter. In processes utilizing this effect, the index change does not depend on the magnitude of the intensity but rather on the spatial modulation of the intensity.

The nonlinear index of refraction has generated significant scientific and technological interest. It has been utilized in or considered for a variety of applications:

- Nonlinear spectroscopy,
- Correcting optical distortions,
- Optical switching,
- Optical logic gates,
- Optical data processing,
- Optical communications,
- Optical limiting,
- Passive laser mode-locking, and
- Waveguide switches and modulators.

These applications derive from several phenomena that have been discovered and are related to the nonlinear refractive index. These occur through a modulation of the phase or amplitude of an intense optical wave propagating through a nonlinear medium.

The nonlinear index of refraction can also produce detrimental effects, such as unwanted phase aberrations or even optical damage. Thus it becomes an important consideration in any experiment or application employing the manipulation of intense optical beams.

This chapter describes many of the physical processes contributing to the nonlinear index of refraction as well as many of the resulting optical phenomena. First, important definitions and notation are given. Then, many of the physical mechanisms producing the nonlinear index are discussed, with formulas given relating the index change to important microscopic and macroscopic parameters. Finally, the formulas for important parameters in nonlinear optical phenomena resulting from the nonlinear index are presented and discussed.

I. DEFINITIONS AND NOTATION

The general dependence of the nonlinear refractive index on intensity can be expressed as

$$n(\mathbf{r}, t) = n_0(\mathbf{r}, t) + \Delta n[I(\mathbf{r}, t)] \quad (1)$$

This equation indicates that the change in the refractive index Δn over its value at low intensities n_0 has a functional dependence on the intensity $I(\mathbf{r}, t)$. This dependence is made more explicit in the following paragraphs. The majority of phenomena considered occur in isotropic materials. Thus, unless stated otherwise, most of the formulas that follow apply to isotropic media.

Kerr-like media. In this case the nonlinear index change can be written as

$$\Delta n = n_2 \langle \mathbf{E}^{(\sim)} \cdot \mathbf{E}^{(\sim)} \rangle \quad (2)$$

where n_2 is called the nonlinear index coefficient, and the angle brackets indicate a time average over at least one optical cycle. It will be remembered that $\mathbf{E}^{(\sim)}$ is the total real optical field, including the rapidly varying oscillatory behavior. These oscillations are too rapid to observe at optical frequencies: only the time averaged effect is observed. Thus the nonlinear index of refraction depends on the slowly time varying complex amplitude $\mathbf{E}(\mathbf{r}, t)$.

At this point, the result of Eq. (2) will depend on the convention used to describe the real field in complex notation. Following the notation for the expansion of the nonlinear polarization as a power series in the electric field, given in Chapter 1, the result is $\Delta n = 2n_2|\mathbf{E}|^2$. Some authors prefer to write the real field as $\mathbf{E}^{(\sim)}(\mathbf{r}, t) = (1/2)\mathbf{E}(\mathbf{r}, t)e^{-i\omega t} + \text{c.c.}$ With this notation, the result of Eq. (2) is $\Delta n = (1/2)n_2|\mathbf{E}|^2$. Both expressions are valid as long as the expression for the field and related expressions, e.g., the intensity, are consistent. For example, for the former case, $I = 2\epsilon_0 n_0 c |\mathbf{E}|^2$, while for the latter, $I = (1/2)\epsilon_0 n_0 c |\mathbf{E}|^2$, with both expressions in SI units. The reader is cautioned to be aware that these conventions are sometimes mixed together in the literature on nonlinear optics. In this book, the former convention will be adhered to unless explicitly stated otherwise.

Since $I \propto |\mathbf{E}|^2$, it is sometimes convenient to express the nonlinear index in terms of intensity. In this case, the index change is expressed by

$$\Delta n = n_2^I I \quad (3)$$

where n_2^I is the nonlinear index intensity coefficient. In the literature this constant is often called γ or sometimes just n_2 . Although it is often possible to distinguish the intensity coefficient from the field-squared coefficient by their context (they also have different physical units), the two will be identified differently here to avoid confusion. The use of γ is avoided since this also is used for other quantities in expressions of the nonlinear index coefficient (see below).

The nonlinear index coefficient is related to the third order nonlinear susceptibility that appears in the expansion to third order of the nonlinear polarization. From Chapter 1, the i th component ($i = x, y, z$) of this third order polarization for a single wave of frequency ω is given by

$$P_i^{(3)}(\omega) = 3\epsilon_0 \sum_{jkl} \chi_{ijkl}^{(3)}(-\omega; \omega, \omega, -\omega) E_j(\omega) E_k(\omega) E_l^*(\omega) \quad (4)$$

This nonlinear polarization leads to a self-induced index change of the type described above in Eq. (2). Another case of importance is that of a strong wave at frequency ω inducing an index change for a weak probe beam at frequency ω' . For this case the third order polarization is written as

$$P_i^{(3)}(\omega') = 6\epsilon_0 \sum_{jkl} \chi_{ijkl}^{(3)}(-\omega'; \omega', \omega, -\omega) E_j(\omega') E_k(\omega) E_l^*(\omega) \quad (5)$$

Note the difference in degeneracy factor since all four frequencies in the argument of the susceptibility are different in Eq. (5). As noted by Boyd [1], the degeneracy factor will also be 6 even if $\omega' = \omega$, as long as there are two physically distinguishable fields present, such as when the weak probe field propagates in a different direction from the pump field.

Recall from Chapter 1 that, from the symmetry properties of an isotropic medium, the only nonzero independent components of the third order susceptibility are χ_{ijij} , χ_{ijji} , and χ_{ijji} with $\chi_{iiii} = \chi_{ijij} + \chi_{ijji} + \chi_{ijji}$ for all i and j . For a single linearly polarized beam or two parallel linearly polarized beams, n_2 is proportional to χ_{iiii} . The existence of off-diagonal terms in the third order susceptibility indicates the possibility of a nonlinearly induced birefringence in isotropic materials through the third order polarization. This will occur for elliptically polarized waves, or for two beams of nonparallel linear polarization.

Expressions that relate n_2 to the third order susceptibility are given in Table 1 in both SI and cgs units. Although for linear polarization n_2 is written in terms of χ_{xxxx} , it could equivalently be expressed in terms of χ_{yyyy} or χ_{zzzz} . It should also be noted that these expressions were developed in an attempt to be consistent with the way the real field is expressed in complex notation for both the nonlinear index and for the third order polarization. As noted above, this can be in terms of either $\mathbf{E}(\mathbf{r}, t)$ or $(1/2)\mathbf{E}(\mathbf{r}, t)$. As long as the field is expressed in the same way for both formulations, i.e., Eqs. (2), (4), and (5), the expressions in Table 1 will be the same. However, some authors choose to expand the nonlinear polarization in $\mathbf{E}(\mathbf{r}, t)$ but express the nonlinear index for a field in terms of $(1/2)\mathbf{E}(\mathbf{r}, t)$. In this latter case, the expressions for n_2 in Table 1 would all be larger by a factor of 4. Finally, for linear polarization note that the nonlinear index change experienced by a weak probe beam of frequency ω' induced by

Table 1 Formulas Relating n_2 and $\chi^{(3)}$ for Isotropic Materials

	SI	cgs
Self-action		
Linear polarization $\Delta n = 2n_2 E ^2$	$n_2 = \frac{3}{4n_0}\chi_{xxxx}^{(3)}(-\omega; \omega, \omega, -\omega)$	$n_2 = \frac{3\pi}{n_0}\chi_{xxxx}^{(3)}(-\omega; \omega, \omega, -\omega)$
	$= \frac{3}{4n_0}(2\chi_{xxyy}^{(3)} + \chi_{xyyx}^{(3)})$	$= \frac{3\pi}{n_0}(2\chi_{xxyy}^{(3)} + \chi_{xyyx}^{(3)})$
Circular polarization $\Delta n_{\pm} = 2n_2^{cir} E_{\pm} ^2$	$n_2^{cir} = \frac{3}{2n_0}\chi_{xxyy}^{(3)}(-\omega; \omega, \omega, -\omega)$	$n_2^{cir} = \frac{6\pi}{n_0}\chi_{xxyy}^{(3)}(-\omega; \omega, \omega, -\omega)$
Elliptical polarization	$\Delta n_+ = \frac{3}{n_0}[\chi_{xxyy}^{(3)} E_+ ^2 + (\chi_{xxyy}^{(3)} + \chi_{xyyx}^{(3)}) E_- ^2]$	$\Delta n_+ = \frac{12\pi}{n_0}[\chi_{xxyy}^{(3)} E_+ ^2 + (\chi_{xxyy}^{(3)} + \chi_{xyyx}^{(3)}) E_- ^2]$
	$\Delta n_- = \frac{3}{n_0}[\chi_{xxyy}^{(3)} E_- ^2 + (\chi_{xxyy}^{(3)} + \chi_{xyyx}^{(3)}) E_+ ^2]$	$\Delta n_- = \frac{12\pi}{n_0}[\chi_{xxyy}^{(3)} E_- ^2 + (\chi_{xxyy}^{(3)} + \chi_{xyyx}^{(3)}) E_+ ^2]$
Cross-action		
Linear polarization $\Delta n(\omega') = 2n_2^{(weak)} E ^2$	$n_2^{(weak)} = \frac{3}{2n_0}\chi_{xxxx}^{(3)}(-\omega'; \omega', \omega, -\omega)$	$n_2^{(weak)} = \frac{6\pi}{n_0}\chi_{xxxx}^{(3)}(-\omega'; \omega', \omega, -\omega)$
Self-action		
Intensity coefficient $\Delta n = n_2^I I$	$n_2^I = \frac{n_2}{\epsilon_0 n_0 c}$	$n_2^I = \frac{4\pi n_2}{n_0 c}$

a strong beam of frequency ω is twice as large as the self-induced index change experienced by the strong beam.

The nonlinear index change for a circularly polarized beam is weaker than that of a linearly polarized beam, as it depends on only one component of the susceptibility tensor. This index change is also the same for both right-handed and left-handed polarizations.

For an arbitrary elliptical polarization, a circular birefringence is induced in the material. In this situation, the index change Δn_+ , experienced by the left circularly polarized component of the field is generally not equal to the index change Δn_- experienced by the right circularly polarized component. This leads to an intensity induced rotation of the polarization ellipse of the beam. These changes are not conveniently expressed in terms of an n_2 but rather in terms of the third order susceptibility components.

The nonlinear index intensity coefficient is directly related to n_2 in the same way that the intensity is related to the square of the modulus of the field. Table 1 also gives these relations in both SI and cgs units.

Non-Kerr-like media. In Kerr-like media the nonlinear index change exhibits a linear dependence on the optical intensity. In some materials, the dependence is not as simple. Generally, it can be stated that the index change is some function of the intensity, i.e.,

$$\Delta n = f(I) \quad (6)$$

For sufficiently small intensities, depending on the properties of the medium, Eq. (6) can be expanded in a power series of I . The leading term will be linear in I , and thus for sufficiently small intensities, the medium will be Kerr-like and thus the index change can be expressed in terms of an n_2 . For larger intensities this is not possible. An example of this is a medium exhibiting saturable absorption (i.e., where the absorption coefficient of the medium decreases with increasing intensity).

Quasi-Kerr-like media. In these materials it is possible to obtain an expression for the nonlinear index change as a linear function of the intensity. One example was given above for non-Kerr-like materials at low intensities. Other examples exist wherein a physical change in the medium, brought on by the accumulated energy or fluence (integrated intensity) from an optical wave deposited in the medium, causes a change in the index of refraction. In some cases it is possible to express this index change as a linear function of the intensity, which would give the appearance that the medium is Kerr-like. An example of this is a thermal nonlinearity. A further example is that of a cascaded second order effect where there is an intensity dependent shift of the optical phase of a wave. This is possible in, for example, second harmonic generation that is not phase matched. The phase shift can be expressed in terms of an effective index change proportional to intensity, which gives the appearance that the medium is Kerr-like.

Intensity modulation effects. In this case, the index change is not directly dependent on the absolute intensity but rather on the relative modulation of the intensity. The index change can be expressed by

$$\Delta n(\mathbf{r}) \propto \frac{\delta I(\mathbf{r})}{I_0} \quad (7)$$

where I_0 is the total spatially averaged intensity and δI is the spatially modulated part of the intensity, i.e., $I(\mathbf{r}) = I_0 + \delta I(\mathbf{r})$. An example of this type of effect is observed in photorefractive media.

The physical units, in both SI and cgs systems, of several parameters appearing in formulas for the nonlinear index and related phenomena are given in Table 2. SI units are most appropriate for experiments, but cgs units often appear in the literature in theoretical developments and sometimes in experimental data. Sometimes the units are mixed, such as when expressing intensity in W/cm^2 .

Table 2 Physical Units of Important Physical Parameters Utilized in Formulas Related to the Nonlinear Index of Refraction

Physical parameter	SI	cgs
Dipole moment, transition moment	$[\mu] = \text{C-m}$	$[\mu] = \text{sC-cm} = \text{sV-cm}^2$
Linear polarizability	$[\alpha] = \text{C-m}^2/\text{V}$	$[\alpha] = \text{cm}^3$
Second hyperpolarizability	$[\gamma] = \text{C-m}^4/\text{V}^3$	$[\gamma] = \text{cm}^5/\text{sV}^2$
Third order polarization	$[P^{(3)}] = \text{C/m}^2$	$[P^{(3)}] = \text{sV/cm} = (\text{erg/cm}^3)^{1/2}$
Electric field	$[E] = \text{V/m}$	$[E] = \text{sV/cm} = (\text{erg/cm}^3)^{1/2}$
Third order susceptibility	$[\chi^{(3)}] = \text{m}^2/\text{V}^2$	$[\chi^{(3)}] = \text{cm}^2/\text{sV}^2$ $= \text{cm}^3/\text{erg} = \text{esu}$
Nonlinear index of refraction (field) coefficient	$[n_2] = \text{m}^2/\text{V}^2$	$[n_2] = \text{cm}^2/\text{sV}^2$ $= \text{cm}^3/\text{erg} = \text{esu}$
Nonlinear index of refraction (intensity) coefficient	$[n_2'] = \text{m}^2/\text{W}$	$[n_2'] = \text{cm}^2\text{-s/erg}$
Vacuum permittivity	$[\epsilon_0] = \text{F/m} = \text{C/V-m}$	—
Distance, length	$[z] = [r] = [L] = \text{m}$	$[z] = [r] = [L] = \text{cm}$
Speed of light	$[c] = \text{m/s}$	$[c] = \text{cm/s}$
Wavelength	$[\lambda] = \text{m}$	$[\lambda] = \text{cm}$
Wave vector	$[k] = \text{m}^{-1}$	$[k] = \text{cm}^{-1}$
Absorption coefficient	$[\alpha] = \text{m}^{-1}$	$[\alpha] = \text{cm}^{-1}$
Planck's constant	$[\hbar] = [h/2\pi] = \text{J-s}$	$[\hbar] = [h/2\pi] = \text{erg-s}$
Intensity	$[I] = \text{W/m}^2$	$[I] = \text{erg/s-cm}^2$
Power	$[P] = \text{W}$	$[P] = \text{erg/s}$

Formulas for converting parameters in cgs units to SI units, and for some mixed units, are given in [Table 3](#).

II. PHYSICAL MECHANISMS

Several physical mechanisms that contribute to the nonlinear index of refraction are discussed below. Formulas for n_2 and $\chi^{(3)}$ in terms of other physical parameters for the various mechanisms are summarized in [Tables 4](#) and [5](#), respectively.

A. Electronic Polarization

This mechanism involves a distortion of the electron cloud about an atom or molecule by the optical field. If the atom or molecule is highly polarizable, then it can exhibit a significant electronic nonlinearity.

Table 3 Conversion Formulas Between the SI and cgs Systems of Units for Several Physical Parameters Utilized in Nonlinear Index of Refraction Processes

μ	(SI) = $\frac{1}{3} \times 10^{-11} \mu$	(cgs)
α	(SI) = $\left(\frac{1}{3}\right)^2 \times 10^{-15} \alpha$	(cgs)
γ	(SI) = $\left(\frac{1}{3}\right)^4 \times 10^{-23} \gamma$	(cgs)
$P^{(3)}$	(SI) = $\frac{1}{3} \times 10^{-5} P^{(3)}$	(cgs)
E	(SI) = $3 \times 10^4 E$	(cgs)
$\chi^{(3)}$	(SI) = $\frac{4\pi}{3^2} \times 10^{-8} \chi^{(3)}$	(cgs)
n_2	(SI) = $\left(\frac{1}{3}\right)^2 \times 10^{-8} n_2$	(cgs)
n_2^I	(SI) = $10^3 n_2^I$	(cgs)
n_2^I	(cm ² /W) = $10^4 n_2^I$ (SI) = $10^7 n_2^I$	(cgs)
L	(SI) = $10^{-2} L$ (cgs)	
c	(SI) = $10^{-2} c$	(cgs)
λ	(SI) = $10^{-2} \lambda$	(cgs)
k	(SI) = $10^2 k$	(cgs)
α	(SI) = $10^2 \alpha$	(cgs)
I	(SI) = $10^{-3} I$	(cgs)
I	(W/cm ²) = $10^{-4} I$ (SI) = $10^{-7} I$	(cgs)
$\mathcal{P}$	(SI) = $10^{-7} \mathcal{P}$	(cgs)

The nonlinear susceptibility is related to the second hyperpolarizability, which is a microscopic parameter. The microscopic dipole moment for any material can, under certain circumstances, be expanded in a power series of the local field at the site of the dipole:

$$\mu_p = \mu_p^0 + \sum_q \alpha_{pq} E_q^{\text{loc}} + D^{(2)} \sum_{qr} \beta_{pqr} E_q^{\text{loc}} E_r^{\text{loc}} + D^{(3)} \sum_{qrs} \gamma_{pqrs} E_q^{\text{loc}} E_r^{\text{loc}} E_s^{\text{loc}} \quad (8)$$

where μ_p is the p th component of the total dipole moment, and μ_p^0 is the p th component of the permanent dipole moment, if one exists. The terms under the summation symbols are the various orders of the induced dipole moment, with $D^{(n)}$ the degeneracy for the n th order term. The third order nonlinearity derives from the second hyperpolarizability:

$$\varepsilon_0 \chi_{ijkl}^{(3)}(-\omega; \omega, \omega, -\omega) =$$

$$f^4 \sum_m N_m \left[\sum_{pqrs} \langle (\hat{i} \cdot \hat{p})(\hat{j} \cdot \hat{q})(\hat{k} \cdot \hat{r})(\hat{l} \cdot \hat{s}) \rangle_m \gamma_{pqrs}^m(-\omega; \omega, \omega, -\omega) \right] \quad (9)$$

where m ranges over the number of species making up the material. The angular brackets indicate an orientational average over the direction cosines relating

Table 4 Formulas for n_2 or n_2^I Due to Various Physical Mechanisms^a

Physical mechanism	SI	cgs
Electronic polarization	$n_2 = \frac{3}{4n_0} \chi_{xxxx}^{(3)}(-\omega; \omega, \omega, -\omega)$	$n_2 = \frac{3\pi}{n_0} \chi_{xxxx}^{(3)}(-\omega; \omega, \omega, -\omega)$
Raman-induced Kerr effect [g_s = Stokes gain coefficient]	$ n_2 \simeq \frac{\epsilon_0 n_0 c \lambda_s}{8\pi} g_s$	$ n_2 \simeq \frac{n_0 c \lambda_s}{32\pi^2} g_s$
Molecular reorientation $f = (n_0^2 + 2)/3$		
No axial symmetry	$n_2 = \frac{f^4 N}{\epsilon_0 n_0} \left[\frac{(\alpha_{\xi\xi} - \alpha_{\eta\eta})^2 + (\alpha_{\eta\eta} - \alpha_{\zeta\zeta})^2 + (\alpha_{\xi\xi} - \alpha_{\zeta\zeta})^2}{90 k_B T} \right]$	$n_2 = \frac{4\pi f^4 N}{n_0} [(\alpha_{\xi\xi} - \alpha_{\eta\eta})^2 + (\alpha_{\eta\eta} - \alpha_{\zeta\zeta})^2 + (\alpha_{\xi\xi} - \alpha_{\zeta\zeta})^2] / 90 k_B T$
Axial symmetry	$n_2 = \frac{f^4 N}{\epsilon_0 n_0} \left[\frac{(\alpha_{\parallel} - \alpha_{\perp})^2}{45 k_B T} \right]$	$n_2 = \frac{4\pi f^4 N}{n_0} \left[\frac{(\alpha_{\parallel} - \alpha_{\perp})^2}{45 k_B T} \right]$
Axial symmetry (pretransitional behaviour)	$n_2 = \frac{f^4 N}{\epsilon_0 n_0} \left[\frac{(\alpha_{\parallel} - \alpha_{\perp})^2}{9a(T - T^*)} \right]$	$n_2 = \frac{4\pi f^4 N}{n_0} \left[\frac{(\alpha_{\parallel} - \alpha_{\perp})^2}{9a(T - T^*)} \right]$
Axial symmetry (homeotropically aligned liquid crystal cell)	$n_2^I(z) = \frac{(\Delta\epsilon/\epsilon_0)^2 \sin^2 2\beta}{4Kc} (Lz - z^2)$ $\langle n_2^I \rangle = \frac{(\Delta\epsilon/\epsilon_0)^2 L^2 \sin^2 2\beta}{24Kc}$	$n_2^I(z) = \frac{(\Delta\epsilon)^2 \sin^2 2\beta}{4Kc} (Lz - z^2)$ $\langle n_2^I \rangle = \frac{(\Delta\epsilon)^2 L^2 \sin^2 2\beta}{24Kc}$
Electrostriction	$n_2 = \frac{\epsilon_0}{36v_a^2 \rho} \frac{[(n_0^2 + 2)(n_0^2 - 1)]^2}{n_0}$	$n_2 = \frac{1}{144\pi v_a^2 \rho} \frac{[(n_0^2 + 2)(n_0^2 - 1)]^2}{n_0}$
Homogeneously broadened two-level system steady state	$n_2 = \frac{\alpha_{00}}{4(\omega_{eg}/c)n_0} \left[\frac{T_2 \Delta}{(1 + T_2^2 \Delta^2)^2} \right] \frac{1}{ E_s^0 ^2}$	$n_2 = \frac{\alpha_{00}}{4(\omega_{eg}/c)n_0} \left[\frac{T_2 \Delta}{(1 + T_2^2 \Delta^2)^2} \right] \frac{1}{ E_s^0 ^2}$

(continued)

Table 4 Continued

Physical mechanism	SI	cgs
Homogeneously broadened two-level system	$n_2 = \frac{1}{2n_0} \frac{N \mu_{eg} ^4}{\epsilon_0(\hbar\Delta)^3}$	$n_2 = \frac{2\pi N \mu_{eg} ^4}{n_0(\hbar\Delta)^3}$
Adiabatic following		
Semiconductors		
Free carrier absorption (m_e = effective mass)	$n_2^f = -\frac{e^2\tau_R\alpha_0}{2m_e\epsilon_0n_0\hbar\omega^3}$	$n_2^f = -\frac{2\pi e^2\tau_R\alpha_0}{m_en_0\hbar\omega^3}$
Band filling	$n_2^f = \left[\frac{d(\Delta n)}{dI} \right]_{I=0}$ $\Delta n(\omega, I) = \frac{c}{\pi} \times \text{P.V.} \int_0^\infty \frac{\Delta\alpha(\omega', I)}{(\omega')^2 - \omega^2} d\omega'$	$n_2^f = \left[\frac{d(\Delta n)}{dI} \right]_{I=0}$ $\Delta n(\omega, I) = \frac{c}{\pi} \times \text{P.V.} \int_0^\infty \frac{\Delta\alpha(\omega', I)}{(\omega')^2 - \omega^2} d\omega'$
Thermal	$n_2^f = \frac{\alpha\tau}{\rho C} \frac{dn}{dT}$	$n_2^f = \frac{\alpha\tau}{\rho C} \frac{dn}{dT}$
Cascaded SHG	$n_2^f(\omega) = \frac{4\pi d_{\text{eff}}^2}{\epsilon_0 n_\omega^2 n_{2\omega} c \lambda_\omega \Delta k}$	$n_2^f(\omega) = \frac{256\pi^4 d_{\text{eff}}^2}{n_\omega^2 n_{2\omega} c \lambda_\omega \Delta k}$
Photorefractive effect (sinusoidal index modulation due to two interfering waves)	$\Delta n(x) = -n_0^3 r_{\text{eff}} \left(\frac{E_D}{1 + E_D/E_N} \right) \times \frac{\sqrt{I_1 I_2}}{I_1 + I_2} \sin Kx$ $E_D = \frac{k_B T K}{e}$ $E_N = \frac{e N_A}{\epsilon_{dc} K}$	

^a Parameters are defined in the text.

Table 5 Formulas for $\chi^{(3)}$ in SI Units Due to Various Physical Processes^a

Physical mechanism	
Electronic polarization	$\chi_{iiii}^{(3)}(-\omega; \omega, \omega, -\omega) = \frac{f^4 N}{\epsilon_0} \gamma_{iiii}(-\omega; \omega, \omega, -\omega)$ $\gamma_{iiii} \equiv \gamma$ $\gamma_{ijij} = \gamma_{ijji} = \gamma_{jiij} = \frac{1}{3} \gamma$
Molecular reorientation	$\chi_{ijkl}^{(3)}(-\omega; \omega, \omega, -\omega) = \frac{f^4 N}{\epsilon_0} [3(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl}) - 2\delta_{ij}\delta_{kl}]$ $\times \left[\frac{(\alpha_{\xi\xi} - \alpha_{\eta\eta})^2 + (\alpha_{\eta\eta} - \alpha_{\zeta\zeta})^2 + (\alpha_{\xi\xi} - \alpha_{\zeta\zeta})^2}{270k_B T} \right]$ $\chi_{ijij}^{(3)} = \chi_{ijji}^{(3)} = \frac{f^4 N}{\epsilon_0} \left[\frac{(\alpha_{\xi\xi} - \alpha_{\eta\eta})^2 + (\alpha_{\eta\eta} - \alpha_{\zeta\zeta})^2 + (\alpha_{\xi\xi} - \alpha_{\zeta\zeta})^2}{90k_B T} \right]$ $\chi_{ijji}^{(3)} = -\frac{f^4 N}{\epsilon_0} \left[\frac{(\alpha_{\xi\xi} - \alpha_{\eta\eta})^2 + (\alpha_{\eta\eta} - \alpha_{\zeta\zeta})^2 + (\alpha_{\xi\xi} - \alpha_{\zeta\zeta})^2}{135k_B T} \right]$
Electrostriction	$\chi_{iiii}^{(3)} = \frac{1}{\epsilon_0} \frac{\gamma_e}{v_a^2} \frac{\partial \epsilon}{\partial \rho} = \frac{\epsilon_0}{27v_a^2 \rho} (n_0^2 + 2)^2 (n_0^2 - 1)^2$
Homogeneously broadened two-level system Steady state	$\chi(E ^2) = \frac{\alpha_{00}}{\omega_{eg}/c} \frac{-T_2\Delta + i}{1 + (T_2\Delta)^2 + E ^2/ E_s^0 ^2}$ $\chi^{(1)} = \frac{\alpha_{00}}{\omega_{eg}/c} \frac{-T_2\Delta + i}{1 + (T_2\Delta)^2}$ $\chi^{(3)} = \frac{\alpha_{00}}{3\omega_{eg}/c} \frac{T_2\Delta - i}{[1 + (T_2\Delta)^2]^2} \frac{1}{ E_s^0 ^2} = -\frac{\chi^{(1)}}{3 E_s^0 ^2}$
Adiabatic following	$\chi^{(3)} = \frac{2N \mu_{eg} ^4}{3\epsilon_0(\hbar\Delta)^3}$

^a Parameters are defined in the text.

the atomic or molecular coordinates $\{\xi\eta\zeta\}$ to the laboratory coordinates $\{xyz\}$; γ_{pqrs}^m is the second hyperpolarizability for species m , N_m is the number of atomic or molecular units of species m per unit volume, and f is a local field factor.

For a centrosymmetric material, the third order term is the lowest order nonlinearity. If the atomic or molecular unit is symmetric, it has no preferred direction of axes. Hence the laboratory system, defined by the vector properties of the applied fields, determines the coordinate system of the microscopic system. The direction cosines are thus all equal to 1, and for a single species Eq. (9) reduces to the expression given in Table 5. The local field factor is then simply

$$f = \frac{n_0^2 + 2}{3} \quad (10)$$

where again n_0 is the linear index of refraction.

The second hyperpolarizability can be computed from quantum mechanics using perturbation theory. The full expression contains 48 terms, all related to a product of four transition dipole moment matrix elements (in the numerators) and energy level transition frequencies (in the denominators). This full expression is not in itself particularly useful for calculations since, in general, the matrix elements and the transition frequencies (i.e., electron energy levels) must further be computed in terms of the basis set of states of the system. This is not a trivial problem, and various *ab initio* and semiempirical methods have been developed to make these calculations [2,3]. The calculations are numerical in nature, since no simple analytical formulas can be found, and they generally require powerful computational methods and long computation times. These computational methods are beyond the scope of this book, and the reader is referred to the literature for further details [2].

When the optical frequency is close to a single or two-photon resonance, then a few terms dominate the contribution to the second hyperpolarizability. Boyd [1] gives an excellent discussion of these terms. Under these conditions, the single beam n_2 is due primarily to the second hyperpolarizability given by

$$\gamma_{iiii}(-\omega; \omega, \omega, -\omega) = \frac{2}{3\hbar^3} \left[\sum'_{abc} \frac{\mu_{gc}^i \mu_{cb}^i \mu_{ba}^i \mu_{ag}^i}{(\omega_{cg} - \omega)(\omega_{bg} - 2\omega)(\omega_{ag} - \omega)} - \sum_{ac} \frac{\mu_{gc}^i \mu_{cg}^i \mu_{ga}^i \mu_{ag}^i}{(\omega_{cg} - \omega)(\omega_{ag} - \omega)(\omega_{ag} - \omega)} \right] \quad (11)$$

where the sums are over a complete basis set of the system, with g representing the ground state and a, b, c representing higher lying excited states. The prime on the first summation signifies that $b = g$ is excluded from the sum. The μ_{ab}^i are the i th components of the transition moment matrix element between states a and b, etc., and $\omega_{ag} = \omega_a - \omega_g$ is the transition frequency between states a and g, etc., where $\hbar\omega_a$ is the energy level of state a, and similarly for other states. It can be seen that the first term contains a two-photon resonance, whereas the second term contains only single photon resonances.

As discussed by Boyd [1], when ω is smaller than any resonance of the system, the two-photon resonant term tends to give rise to a positive n_2 , since the linear polarizability of excited states tend to be larger than that of the ground state (i.e., the electrons spend more time further away from the nuclei). On the other hand, a single photon resonance will generally give rise to a negative n_2 , since a single photon resonance will cause a partial depletion of the ground state leading to a decrease in the molecular polarizability (see Section II.E).

For nonresonant electronic processes in transparent dielectrics, a typical value for the second hyperpolarizability is $\gamma \sim 10^{-36}$ esu $\sim 10^{-61}$ C-m⁴/V³,

although large organic molecules can have $\gamma \sim (100\text{'s to } 1000\text{'s}) \times 10^{-36}$ esu. Typically, $\chi^{(3)} \sim 10^{-14}$ esu $\sim 10^{-22}$ m²/V², and $n_2 \sim 10^{-13}$ esu $\sim 10^{-22} - 10^{-21}$ m²/V². Some polymeric systems have been developed with nonresonant $\chi^{(3)} \sim 10^{-12} - 10^{-10}$ esu $\sim 10^{-20} - 10^{-18}$ m²/V². Nonresonant electronic processes are very fast, with typical time responses $\sim 10^{-15}$ s.

B. Raman Induced Kerr Effect

This effect is related to stimulated Raman scattering (see Chapter 14) and is an example of a strong beam inducing a refractive index change for a weak probe beam. The strong beam incident on a Raman active medium is from a laser at frequency ω_L . A weak probe beam of frequency ω_S is also incident on the medium. The weak beam is referred to as the Stokes beam with $\omega_S < \omega_L$.

Linear Raman scattering is a two-photon process wherein a laser photon scatters into a Stokes photon, which is down-shifted in frequency from the laser frequency by an amount equal to a Raman active mode in the medium, e.g., a molecular vibrational quantum (see Fig. 1). Stimulated Raman scattering is a four-photon process wherein a Stokes beam incident on the Raman active medium is amplified. The gain in the Stokes beam is at the expense of photons lost from the laser beam. However, for a strong laser beam and weak Stokes beam, the laser field can be treated as approximately constant (nondepleted pump approximation).

Stimulated Raman scattering can be described as a third order nonlinear process. The third order polarization at the Stokes frequency is given by

$$P_S^{(3)} = 6\epsilon_0\chi_R^{(3)}(-\omega_S; \omega_S, \omega_L, -\omega_L)E_S|E_L|^2 \quad (12)$$

where all fields are linearly polarized and are parallel. The subscript R signifies a Raman susceptibility, which is a complex quantity. The Raman gain factor is

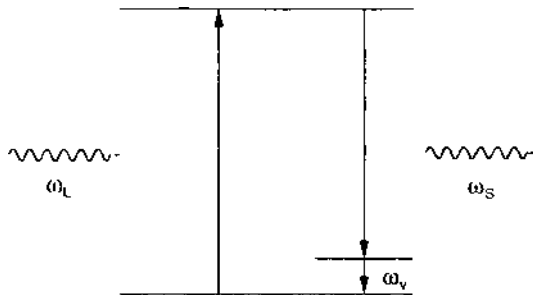


Figure 1 Schematic diagram of Raman scattering.

related to the imaginary part of this susceptibility:

$$G(\omega_S) = -\frac{6\omega_S}{n_S c} \text{Im}[\chi_R^{(3)}(-\omega_S; \omega_S, \omega_L, -\omega_L)]|E_L|^2 \quad (13)$$

where n_S is the linear index of refraction at the Stokes frequency, and the exponential gain factor $G(\omega_S)$ has units of m^{-1} . The real and imaginary parts of $\chi_R^{(3)}$ are related through a Kramers–Kronig relation.

The frequency dependence of the gain factor displays a Raman resonance:

$$G(\omega_S) = \frac{(\Gamma/2)^2 G^{\text{peak}}}{[\omega_S - (\omega_L - \omega_V)]^2 + (\Gamma/2)^2} \quad (14)$$

where G^{peak} is the peak gain, Γ is the Raman linewidth (FWHM), and $\omega_L - \omega_V$ is the Raman resonance. The nonlinear index coefficient is related to the real part of $\chi_R^{(3)}$ and can be expressed in terms of the gain coefficient g_s , which is the gain factor $G(\omega_S)$ per unit (laser) intensity. When the probe (Stokes) frequency ω_S is within $\pm \Gamma/2$ of resonance, the approximate relation shown in Table 5 is obtained. Note that $\text{Re}[\chi_R^{(3)}]$ will change sign as the Stokes frequency is swept through resonance.

For typical values of $g_s \sim 3 \times 10^{-11} \text{ m/W}$, $n_L \sim n_S \sim 1.5$, $\lambda_S \sim 10^{-6} \text{ m}$, the real part of the third order Raman susceptibility has a typical value of $|\text{Re}[\chi_R^{(3)}]| \sim 10^{-20} \text{ m}^2/\text{V}^2$. This is about two orders of magnitude larger than typical nonresonant electronic processes because of the resonant nature of the Raman effect. The response times of these effects are of the order of the lifetime of the Raman induced material excitation (e.g., a higher lying vibrational state in the ground electronic manifold of states of a molecule), and is typically $\sim 10^{-12} \text{ s}$.

C. Molecular Orientational Effects

Anisotropic molecules (i.e., displaying an anisotropic linear polarizability tensor) will tend to exhibit an optically isotropic behavior in the bulk when they are disordered, i.e., when their orientations are randomly distributed. This is true of several liquids (e.g., CS_2) and of liquid crystals above the transition to a mesoscopic phase.

When a strong electric field is applied to such a system, the induced dipole moments of the molecules experience a torque attempting to align the most polarizable axis with the applied field, working against the thermal fluctuation forces (i.e., molecular collisions). An incident optical wave polarized along the direction of the strong field will experience an increase in the local index of refraction, since it sees, on the average, more highly polarizable molecules than when the strong alignment field is absent. The situation is depicted schematically in Fig. 2.

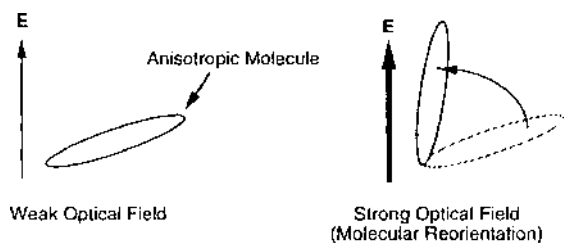


Figure 2 Anisotropic molecular reorientation by a strong electric field.

When the applied field is dc, this effect is called the Kerr effect. Similarly, the field could be due to a strong optical wave. The system then responds not to the instantaneous field but to the time averaged field squared, as in Eq. (1), and the effect is then termed the ac or optical Kerr effect. When the alignment field and the incident optical field are one and the same, this leads to a self-induced refractive index change expressible by an n_2 as long as the induced dipole–electric field interaction energy is small compared to the thermal energy, which is typically the case.

The induced dipole moment of an anisotropic molecule can be expressed in terms of the principal polarizability components corresponding to the principal axes of the molecule: $\alpha_{\xi\xi}$, $\alpha_{\eta\eta}$, and $\alpha_{\zeta\zeta}$. When a strong field is applied to such a system, the system will attempt to minimize the electrical energy $(-\frac{1}{2}\int \mathbf{\mu} \cdot d\mathbf{E}^{(-)})$ by aligning the most polarizable axis (usually designated by the ζ -axis) to the field. The result is a new thermal equilibrium state of the system. The average polarizability of the medium in this new state can be calculated assuming a Boltzmann distribution of molecular orientations. To lowest order in $|\mathbf{E}|^2$, the average polarizability is given by a linear term $\langle\alpha_0\rangle = \frac{1}{3}(\alpha_{\xi\xi} + \alpha_{\eta\eta} + \alpha_{\zeta\zeta})$ plus a nonlinear term proportional to $|\mathbf{E}|^2$. The nonlinear term can thus be expressed in terms of a third order susceptibility, which is given in Table 5.

For axially symmetric molecules (e.g., CS_2), $\alpha_{\xi\xi} = \alpha_{\eta\eta} \equiv \alpha_{\perp}$ and $\alpha_{\zeta\zeta} \equiv \alpha_{\parallel}$. Then writing $\Delta\alpha = \alpha_{\parallel} - \alpha_{\perp}$, the factor containing the molecular polarizability components becomes

$$\frac{(\alpha_{\xi\xi} - \alpha_{\eta\eta})^2 + (\alpha_{\eta\eta} - \alpha_{\zeta\zeta})^2 + (\alpha_{\xi\xi} - \alpha_{\zeta\zeta})^2}{270k_B T} \rightarrow \frac{(\Delta\alpha)^2}{135k_B T} \quad (15)$$

where k_B is Boltzmann's constant ($1.38 \times 10^{-23} \text{ J/K} = 1.38 \times 10^{-16} \text{ erg/K}$), and T is the temperature in kelvins (K).

The usual self-induced nonlinear index coefficient n_2 is given in terms of $\chi_{iiii}^{(3)}(-\omega; \omega, \omega, -\omega)$ where $i = x, y$, or z . For this so-called degenerate case,

$$\chi_{ijij}^{(3)}(-\omega; \omega, \omega, -\omega) = \chi_{ijij}^{(3)}(-\omega; \omega, \omega, -\omega) \quad (16)$$

for all i, j , which can be seen to follow from the intrinsic permutation symmetry of the nonlinear susceptibility (see Chapter 1), and

$$\chi_{iji}^{(3)}(-\omega; \omega, \omega, -\omega) = 6\chi_{ijj}^{(3)}(-\omega; \omega, \omega, -\omega) \quad (17)$$

for all i, j for molecular reorientation processes.

The situation is somewhat different for the case of a strong pump induced nonlinear index for a weak probe. In this case the susceptibility $\chi_{iii}^{(3)}(-\omega'; \omega', \omega, -\omega) = \chi_{ijj}^{(3)}(-\omega'; \omega', \omega, -\omega) + \chi_{iji}^{(3)}(-\omega'; \omega', \omega, -\omega) + \chi_{ijj}^{(3)}(-\omega'; \omega', \omega, -\omega) = \chi_{iii}^{(3)}(-\omega, \omega, \omega, -\omega)$, i.e., it is equal to its degenerate counterpart, ignoring dispersion, but the relations between its three nonzero components is different from those of the degenerate case:

$$\begin{aligned} \chi_{iji}^{(3)}(-\omega'; \omega', \omega, -\omega) &= \chi_{ijj}^{(3)}(-\omega'; \omega', \omega, -\omega) \\ &= -\frac{2}{3}\chi_{ijj}^{(3)}(-\omega'; \omega', \omega, -\omega) \end{aligned} \quad (18)$$

The significance of this is as follows. As stated earlier, the nonlinear index coefficient for the weak probe, when the pump and probe are copolarized, is twice as large as the self-induced index coefficient for the pump, i.e.,

$$(n_2^{(\text{weak})})_{ii} = 2n_2 \quad (19)$$

while the index coefficient for a cross-polarized pump and probe is

$$(n_2^{(\text{weak})})_{ij} = -\frac{1}{2}(n_2^{(\text{weak})})_{ii} \quad (20)$$

These equations are true for the nondegenerate case as well as the degenerate case when the pump and probe are physically distinguishable fields. The situation in Eq. (20) arises for molecular reorientation processes because the strong pump tends to align molecules along its polarization axis, while the probe samples the index perpendicular to this direction. Hence it sees an index reduced from the no-pump situation since, on the average, the molecules it samples have a smaller polarizability. This is depicted schematically in Fig. 3. Note that in a molecular reorientation process, the net polarizability of the medium has not increased. The optical field merely rearranges the polarizability present among the various tensor components.

Typical nonlinearities associated with small anisotropic molecular systems are $\chi^{(3)} \sim 10^{-13} - 10^{-12}$ esu $\sim 10^{-22} - 10^{-21}$ m²/V², and $n_2 \sim 10^{-12} - 10^{-11}$ esu $\sim 10^{-21} - 10^{-20}$ m²/V². The response time is $\sim 10^{-12}$ s. Liquid crystals in their isotropic phase will exhibit larger nonlinearities but with slower response times. These materials are particularly interesting in their mesoscopic phase or near the transition to such a phase.

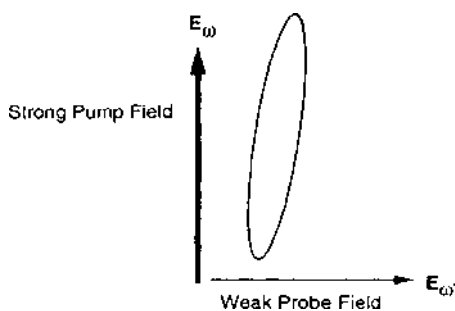


Figure 3 Molecular reorientation by a strong pump field probed with a weak orthogonally polarized field.

A system of axially symmetric liquid crystal molecules will exhibit a pretransitional behavior near its isotropic-to-nematic transition in which the optical nonlinearity can grow very large. The susceptibility is very temperature dependent in this case as shown in Table 5. It can be seen that as the temperature is lowered, the susceptibility increases as $(T - T^*)^{-1}$, where T^* is a fictitious second order phase transition temperature approximately equal to the critical temperature for the isotropic-to-nematic transition.

The parameter a in the expression for the susceptibility is a constant in the expansion of the free energy F of the system in terms of an order parameter Q [3,4,5]:

$$F = F_0 + \frac{1}{2}a(T - T^*)Q^2 + \frac{1}{3}BQ^3 + \dots - \text{const.} \times Q|E|^2 \quad (21)$$

The order parameter is defined by

$$Q = \frac{3}{2} \left\langle \cos^2 \theta - \frac{1}{3} \right\rangle \quad (22)$$

where θ is the angle between the long axis of the molecule and the direction of the optical electric field. The maximum order parameter is 1.

In the nematic phase, liquid crystal molecules are highly correlated and act cooperatively. This is the origin of the large nonlinearity observed in this phase. Pretransitional behavior describes the system optical response as this transition is approached. The apparent divergence in the susceptibility with temperature is a manifestation of the gradual transition from the isotropic to the nematic phase. The nonlinear index of refraction can increase by orders of magnitude as this transition is approached, but the response of the system exhibits a critical slowing

down with a response time given by

$$\tau = \frac{\eta}{a(T - T^*)} \quad (23)$$

where η is the coefficient of viscosity. In the pretransition region, the nonlinear index coefficient can approach $n_2 \sim 10^{-9}$ esu, but the response time is $\sim 10^{-7}$ s.

In the nematic phase, the cooperative behavior of the liquid crystal molecules produces a nonlocal response. This results in a nonlinear index that is a function of position. An example taken from Khoo and Wu [5] is illustrated in Fig. 4. This depicts an optical wave propagating through a homeotropically aligned nematic liquid crystal cell of thickness L . The wave vector of the field makes an angle β with respect to the average liquid crystal director field (the direction of the long molecular axis). The nonlinear index intensity coefficient is given in Table 4. This expression is valid for small angle distortions of the nematic director field. Note that this index coefficient is z dependent, where the z -axis coincides with the average nematic director orientation.

For computational purposes in devices, the average nonlinear index coefficient over the cell thickness is of interest. This is also given in Table 4. Now note that the coefficient varies quadratically with the cell thickness. The anisotropy parameter $\Delta\epsilon$ is the difference in the optical dielectric constants measured parallel to and perpendicular to the long molecular axis. K is a typical elastic restoring force constant for the nematic (the splay constant in this example).

Khoo and Wu [5] give some typical values for the parameters associated with a liquid crystal cell (in cgs units): $\Delta\epsilon = 0.6$, $K = 10^{-6}$ dyn, $L = 10^{-2}$ cm,

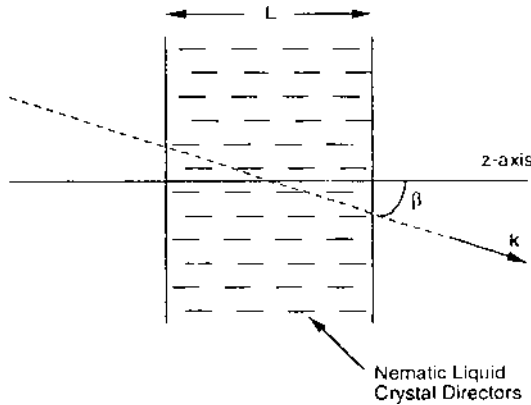


Figure 4 Schematic diagram of a wave propagating through a homeotropically aligned nematic liquid crystal cell.

$\beta = 45^\circ$. These yield an average index coefficient of $\langle n_2^I \rangle = 5 \times 10^{-11} \text{ cm}^2\text{-s/erg} = 5 \times 10^{-4} \text{ cm}^2/\text{W}$. This is about 10 orders of magnitude larger than the n_2^I achieved with small anisotropic molecular systems (e.g., CS_2). However, the response time ($\sim 10^{-3} - 10^{-2} \text{ s}$) is much longer.

The equation given in Table 4 indicates that the nonlinear index change is zero when $\beta = 0$. However, for this case with a homeotropically aligned liquid crystal cell, there is a highly nonlinear refractive index change above a critical intensity. This change is associated with the Fredericks transition. See Ref. 5 for more details on this highly nonlinear phenomenon.

D. Electrostriction

Electrostriction is a phenomenon that depends on the presence of an inhomogeneous intensity, i.e., a spatially varying, time averaged electric field. This would occur, for example, in the superposition of coherent waves to form an interference pattern of bright and dark fringes, or along the transverse direction to a propagating Gaussian shaped beam. Such an inhomogeneous field produces a force on the molecules or atoms comprising a system called the electrostrictive force.

The electrostrictive force is proportional to the gradient of the square modulus of the electric field. The constant of proportionality, $(1/2)\gamma_e$, is given in terms of the electrostrictive coefficient, where

$$\gamma_e = \rho \left(\frac{\partial \epsilon}{\partial \rho} \right) \quad (24)$$

with ρ the mass density and $\epsilon = \epsilon_0 n_0^2$. This force can be understood from the fact that the induced dipoles in the medium will experience a translational force in a nonuniform field that is proportional to the gradient of the field. (Dipoles will not experience such a force in a uniform field, although they may experience a torque). The force is such as to move the dipoles into a region of higher intensity. This produces an increase in local density resulting in an increase of the refractive index locally. Thus even a system of isotropic molecules will be electrostrictive.

The nonlinear index coefficient is proportional to $\chi_{xxxx}^{(3)}$ and is given in Table 4, where v_a is the acoustic velocity in the medium. For an electrostrictive medium, the following properties of the susceptibility hold:

$$\begin{aligned} \chi_{ijj}^{(3)}(-\omega; \omega, \omega, -\omega) &= \chi_{ijj}^{(3)}(-\omega; \omega, \omega, -\omega) \\ &= \frac{1}{2} \chi_{iii}^{(3)}(-\omega; \omega, \omega, -\omega) \\ \chi_{iji}^{(3)}(-\omega; \omega, \omega, -\omega) &= 0 \end{aligned} \quad (25)$$

For typical values given by $v_a \sim 10^4$ m/s, $\rho \sim 10^3$ kg/m³, $n_0 \sim 1.4$, the electrostriction contribution to the nonlinear index coefficient is $n_2 \sim 10^{-19}$ m²/V². This is of the same order as the molecular reorientational contribution. The response time, however, is of the order of 10^{-9} s, i.e., about three orders of magnitude slower than the molecular reorientational response time.

E. Population Redistribution

When the frequency of incident radiation is near a resonant energy transition of an atom or molecule, then real transitions are induced. This means that electrons can occupy real excited states for a finite period of time. This is called a population redistribution. Since the optical polarization is ordinarily determined by the total number of atoms or molecules in the ground electronic state (for low intensity light), this population redistribution produces a change in the index of refraction. This effect can be seen in atomic vapors, molecular gases, liquid solutions of organic molecules, transparent dielectric solids doped with metal ions or color centers, and semiconductors.

In some situations, the near-resonant interaction can be described as if the material had only two energy levels. This is because the resonant interaction between the radiatively connected ground and excited states is so strong that other nonresonant interactions can be ignored. It is assumed that the resonant frequency for a transition from the excited state to a higher lying state is far removed from that of the ground and excited states. The situation of semiconductors, with finite valence and conduction energy bands, is somewhat different depending on the time scale of the optical interaction.

Two-level systems. This deceptively simple looking system is illustrated in Fig. 5. The two levels are the ground (g) and excited (e) states. The resonant transition frequency connecting these states is defined by ω_{eg} . The system is also

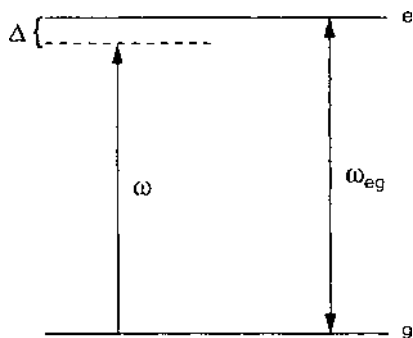


Figure 5 A two-level system.

characterized by a transition dipole moment, μ_{eg} , and an equilibrium population difference $\Delta N^{\text{eq}} = (N_g - N_e)^{\text{eq}}$, where N_g^{eq} and N_e^{eq} are the number densities of atoms or molecules in the ground and excited states, respectively, long after or before the radiation is incident on the medium. The transition dipole moment has a characteristic dephasing lifetime T_2 , which is related to the homogeneous line width (FWHM) of the transition by $\Delta\omega_{\text{FWHM}} = 2\pi/T_2$. A nonequilibrium population difference will decay to its equilibrium value in a time of the order of a characteristic time defined as T_1 .

The average dipole moment of such a system described above is usually derived by density matrix theory. The polarization is then given as the average dipole moment per unit volume. In two particular time regimes, defined as the steady-state and the adiabatic following regimes, the resulting polarization P can be written as a constant times the applied field E . The susceptibility of the system is then given by $\varepsilon_0\chi = P/E$.

The steady-state regime is examined first. Here the optical field has been applied for a time long compared to T_1 and T_2 . Thus all optically induced transient effects have been damped out. This regime is typical of the response of a two-level system to a cw laser.

The applied optical field is close to resonance, and a detuning parameter is defined by

$$\Delta = \omega - \omega_{eg} \quad (26)$$

Since the frequency is near resonance, the susceptibility will be complex, with its real part related to the index of refraction and its imaginary part related to the absorption coefficient. The susceptibility is also proportional to the nonequilibrium population difference, which is itself dependent on the intensity of the incident radiation due to optically induced resonant transitions from the ground to excited states (i.e., absorption). The result is an intensity dependent susceptibility given by

$$\chi = \frac{\alpha_{00}}{\omega_{eg}/c} \frac{i - T_2\Delta}{1 + (T_2\Delta)^2 + |E|^2/|E_s^0|^2} \quad (27)$$

where α_{00} is the ordinary (linear) absorption coefficient at line center ($\Delta = 0$, $|E|/|E_s^0| \ll 1$), which is given by

$$\alpha_{00} = \frac{\omega_{eg} |\mu_{eg}|^2 T_2 \Delta N^{\text{eq}}}{\varepsilon_0 c \hbar} \quad (28)$$

(Equation (28) is valid for an atomic vapor or molecular gas where the linear index of refraction is approximately 1. For molecules in solution or doped in a solid dielectric, the expression for χ should include the local field factors appropriate to the host medium.) A saturation field at line center, determined by

microscopic parameters, is defined by

$$|E_s^0|^2 = \frac{\hbar^2}{4|\mu_{eg}|^2 T_1 T_2} \quad (29)$$

Thus it can be seen that when the applied optical field is small compared to the line center saturation field, the susceptibility is essentially independent of intensity and has a characteristic Lorentzian line shape.

When the applied field is larger, but still small compared to the saturation field, the denominator in Eq. (27) can be expressed as a binomial expansion in terms of the quantity $|E|^2/|E_s^0|^2$. To lowest order in this parameter, the resulting expression for χ has a term independent of the applied field, and a term proportional to $|E|^2$. The former is identified with the linear susceptibility $\chi^{(1)}$. The latter term, through the relationship of the nonlinear polarization, i.e.,

$$P^{(3)} = 3\epsilon_0 \chi^{(3)}(-\omega; \omega, \omega, -\omega) E |E|^2 \quad (30)$$

determines the third order susceptibility $\chi^{(3)}$. These expressions are given in Table 5, and the corresponding expressions for n_2 are in Table 4.

Note that $\chi^{(3)}$ can be expressed compactly in terms of $\chi^{(1)}$ and an off-resonance saturation field defined by

$$|E_s^\Delta|^2 = |E_s^0|^2 [1 + (T_2 \Delta)^2] \quad (31)$$

Furthermore, it is useful to define corresponding saturation intensities by

$$\begin{aligned} I_s^0 &= 2\epsilon_0 n_0 c |E_s^0|^2 \\ I_s^\Delta &= 2\epsilon_0 n_0 c |E_s^\Delta|^2 \end{aligned} \quad (32)$$

Alkali metal vapors exhibit some of the largest resonant nonlinearities. Consider, for example, the $3s \rightarrow 3p$ transition in atomic sodium vapor at a frequency corresponding to a wavelength of 589 nm. The transition dipole moment is approximately $|\mu_{eg}| \approx 2.5ea_0 = 2.0 \times 10^{-29}$ C-m, where a_0 is the Bohr radius. Assuming $N = 10^{20} \text{ m}^{-3}$ and negligible collisional line broadening so that $T_2 \approx 2T_1 = 32 \text{ ns}$, the unsaturated line center absorption coefficient is $\alpha_{00} = 1.5 \times 10^7 \text{ m}^{-1}$. This corresponds to a $1/e$ absorption depth of less than $1 \mu\text{m}$. Let the incident radiation have a frequency below the transition frequency of 1 cm^{-1} . Then the detuning parameter would be $\Delta = -6\pi \times 10^{10} \text{ rad/s}$. From Table 5, the real part of $\chi^{(1)}$ is 2.3×10^{-4} . Hence the linear index of refraction is approximately 1, and local field factors can be ignored. The line center saturation field is then approximately 100 V/m , corresponding to a line center saturation intensity of only 70 W/m^2 . The off-resonance saturation intensity corresponding to this detuning is $36 \text{ MW/m}^2 = 3.6 \text{ kW/cm}^2$. Finally, the value of $\chi^{(3)}$ is then

$-1.6 \times 10^{-16} \text{ m}^2/\text{V}^2$, which corresponds to $n_2 = -1.2 \times 10^{-16} \text{ m}^2/\text{V}^2$. The nonlinear index coefficient is negative because saturation causes the index to decrease for this detuning, as illustrated in Fig. 6.

The magnitude of this n_2 is 4 or 5 orders of magnitude larger than those corresponding to molecular reorientation, but the response time is $\sim T_2$, which for the alkali metal vapors in the noncollisionally line broadened regime is tens of nanoseconds. The effective T_2 can be reduced by introducing a foreign gas (e.g., a noble gas), but this would also raise the saturation intensity. For atoms of comparable dipole moment (i.e., oscillator strength) doped in liquid solutions or solids, the nonlinear index coefficient will be smaller because of the high damping rates induced by molecular or phonon collisions. This reduces the effective T_2 and increases the saturation intensity.

The adiabatic following regime applies to short pulses rather than cw laser beams. It is assumed that the pulse width τ_p satisfies the inequality $\tau_p \ll T_1, T_2$. Also, the detuning is assumed to be such that $|\Delta| \gg T_2^{-1}, \tau_p^{-1}, |\mu_{eg}| |E|/\hbar$. In other words, the detuning is large compared to the homogeneous line width, any Fourier component of the pulse, and the so-called Rabi frequency $\Omega = |\mu_{eg}| |E|/\hbar$ (which leads to power broadening of the line width). The polarization

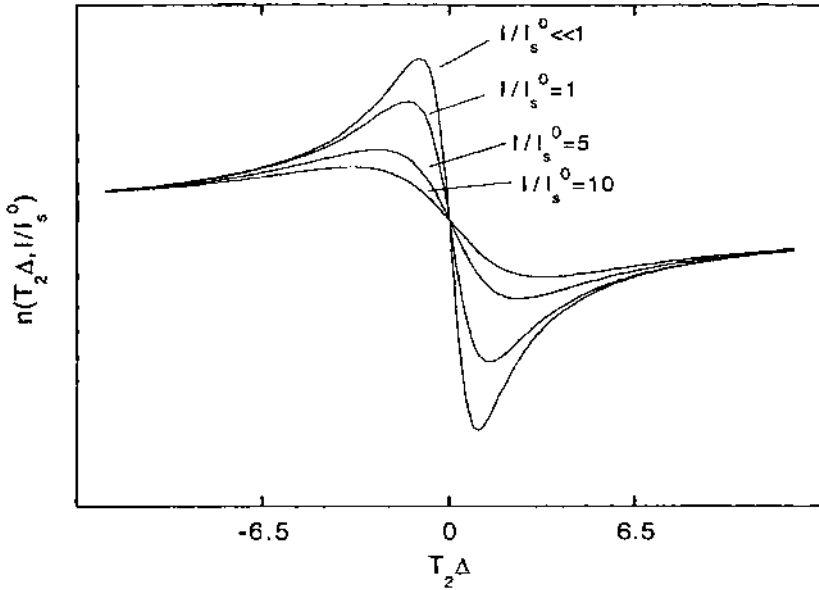


Figure 6 Refractive index of a two-level system as a function of normalized detuning from line center, parameterized by the ratio of incident intensity to the saturation intensity.

can then be written in terms of the Rabi frequency as

$$P = -\frac{\Omega\Delta}{2\Delta^2} \frac{N|\mu_{eg}|}{(1 + |\Omega|^2/\Delta^2)^{1/2}} \quad (33)$$

Expanding the denominator again leads to a third order term that yields the expression for $\chi^{(3)}$ given in [Table 5](#).

Note that the polarization is real in the adiabatic following regime. This is because of the conditions assumed above, which imply that the pulse is far enough from resonance that negligible absorption results, and the pulse is so short that no atomic or molecular relaxation (i.e., damping) can occur for the duration of the pulse. The nonlinear response of the medium then adiabatically follows the optical pulse in time.

Semiconductors. These materials have proved to be very rich and fruitful media for nonlinear optical research and potential applications. They exhibit bound electronic nonlinear effects like wide gap dielectrics (these can be described by the equations in Section II.A), but some of the larger effects are due to electron population redistribution.

One of the main differences in semiconductors over transparent dielectrics is that semiconductors can have a small population of free carriers at thermal equilibrium. These carriers can absorb electromagnetic radiation as they scatter from the lattice. Accompanying this free carrier absorption is a free carrier index of refraction. The refractive index depends directly on the plasma frequency, which itself is a function of the free carrier density. Linear absorption of a photon of energy $\hbar\omega > \mathcal{E}_g$, where $\mathcal{E}_g$ is the energy gap between the valence and conduction bands, promotes electrons to the conduction band, increasing the electron population density. Hence the plasma frequency is modified, producing an intensity dependent change in the refractive index. The resulting nonlinear index coefficient is given in [Table 4](#).

The ω^{-3} dependence makes this effect particularly strong in the infrared. Typical values are $n_2^I \sim 10^{-6} \text{ cm}^2/\text{W}$. Note that α is the linear absorption coefficient across the energy gap, m_e the electron effective mass, and $\tau_R \sim 10 \text{ ns}$ is the electron–hole recombination time.

Linear absorption across the energy gap will produce a redistribution of electron and hole densities. This causes a change in the linear absorption $\Delta\alpha(\omega, I)$ which is intensity dependent. Two time regimes are of interest. For pulses short compared to the intravalley scattering time, the system will respond just like an ensemble of two level absorbers. These cases can be treated by the theory for two-level systems given earlier. This phenomenon is called state filling. It may be observed when subpicosecond pulses are used to excite the semiconductor. For pulses long compared to the intravalley scattering time, the nonlinear effects are due to band filling.

The intensity dependent absorption change is not easily computed for the band filling regime. However, it is readily measured as a function of frequency, or photon energy. Then, as shown in Table 4, the index change can be computed by the Kramers–Kronig relation. This relates $\Delta\alpha(\omega, I)$ to $\Delta n(\omega, I)$ through the integrals given in Table 4, where P.V. stands for the Cauchy principal value of the integral defined by

$$\text{P.V.} \int_A^B f(x)dx = \lim_{\varepsilon \rightarrow 0} \left[\int_A^{a-\varepsilon} f(x)dx + \int_{a+\varepsilon}^B f(x)dx \right] \quad (34)$$

where a is a singularity of the function $f(x)$ on the real axis. The nonlinear index coefficient is given by the derivative of $\Delta n(\omega, I)$ with respect to intensity I in the limit when $I \rightarrow 0$.

Butcher and Cotter [6] outline a model for computing $\Delta\alpha(\omega, I)$ for direct gap group IV and III–V semiconductors. The free electron density is determined by a simple rate equation:

$$\frac{dN_e}{dt} = \frac{\alpha I}{\hbar\omega} - \frac{(N_e - N_e^{\text{eq}})}{\tau_R} \quad (35)$$

The resulting electron density (which is intensity dependent) changes the Fermi levels of the conduction and valence bands. This in turn changes the absorption coefficient and refractive index. The problem must be solved self-consistently. The intensity scale for significant changes in the linear optical properties is determined by the saturation intensity given by

$$I_s = \frac{\varepsilon_0 n_0 c m_e \mathcal{E}_g \Gamma_{cv}}{e^2 \tau_R} \quad (36)$$

where Γ_{cv} is the transition line width and e is the electron charge. Typical values of I_s are on the order of 10^3 W/cm^2 . With $\tau_R \sim 10\text{--}50 \text{ ns}$, typical nonlinear index coefficients for the band filling regime in direct gap semiconductors are $n_2^I \sim 10^{-6}\text{--}10^{-4} \text{ cm}^2/\text{W}$.

The Coulomb interaction between an excited electron and hole can strongly modify the absorption edge of a semiconductor. When the electron and hole form a bound state known as an exciton, an additional line spectrum appears for $\hbar\omega < \mathcal{E}_g$. The photogeneration of free carriers results in an electric field that screens (i.e., reduces the effectiveness of) the field binding the exciton. This strongly saturates the exciton absorption, producing a large change in the refractive index. Formulas for the resulting n_2^I are the same as for the band filling effect, assuming that the intensity dependent change in absorption coefficient as a function of frequency or photon energy can be measured. Butcher and Cotter [6] also give the outline of a theory for computing this exciton saturation effect. The results must be obtained numerically. Typical values are $n_2^I \sim 10^{-6} \text{ cm}^2/\text{W}$.

The above discussion has related the properties of bulk semiconductors. Large nonlinear effects have also been observed in artificially produced semiconductors such as multiple quantum wells and quantum dots. Quantum well materials consist of multiple alternating thin layers (~ 10 nm) of different semiconductor materials. In the thin dimension, quantum confinement effects produce exciton resonances that are sharper than the corresponding ones in bulk semiconductors and are more easily saturated. This results in large optical nonlinearities. Nonlinear index coefficients as large as $n_2^I = -2 \times 10^{-4} \text{ cm}^2/\text{W}$ have been measured. Again, the equations displayed for band filling effects can be used to compute n_2^I if the nonlinear absorption can be measured as a function of frequency. The recovery time of the nonlinear index in quantum wells can be of the order of tens of nanoseconds, or ~ 100 ps for proton-bombarded materials.

Quantum dots are small semiconductor particles with dimensions of a few nanometers. Quantum confinement produces a large separation in energy states with the result that state filling effects dominate the optical nonlinearities. Thus the system is treated as a two-level system as described above. Typical experiments have been performed on semiconductor doped glasses that have a range of particle sizes. This produces an inhomogeneous broadening of the absorption lines with a much higher saturation intensity than homogeneous broadened systems. However, typical measured values give $n_2^I \sim 10^{-12} - 10^{-11} \text{ cm}^2/\text{W}$ with recovery times ~ 10 ps. This fast recovery time is believed to be related to the small size of the semiconductor particles (e.g., surface state effects or large free carrier capture cross-sections by deep lying traps). The development of smaller, more uniformly sized particles with lower saturation intensities promises higher nonlinearities with fast recovery rates.

The nonlinear optics of metallic quantum dots has also been studied. There are no simple formulas for computing the nonlinear index coefficients. The metal spheres support a surface plasmon mode. The nonlinear susceptibility is enhanced when the frequency of the incident optical intensity approaches the resonant frequency of the surface plasmon. Colloidal silver and gold particles have been studied experimentally. Typical values are $\chi^{(3)} \sim 10^{-9}$ to $10^{-8} \text{ esu} \sim 10^{-17}$ to $10^{-16} \text{ m}^2/\text{V}^2$.

F. Thermal Contributions

The nonlinear index intensity coefficient due to thermal contributions in the steady state is shown in Table 4. The change in index due to a temperature rise ΔT can be expressed by

$$\Delta n = \frac{dn}{dT} \Delta T \quad (37)$$

where dn/dT is called the thermooptic coefficient. In most liquids and solids it is

due to a density change (i.e., expansion) with temperature. Since the density decreases in expansion and the refractive index is proportional to density, this contribution to dn/dT is generally negative. In some semiconductors, the absorption band edge will red-shift with temperature (Franz–Keldish effect). This produces a positive thermooptic coefficient.

In the formula given in Table 4, α is the linear absorption coefficient. This could be an intrinsic absorption of the material, or absorption due to impurities. However, any material that has some appreciable absorption will exhibit a thermal nonlinear index of refraction in the steady state. The time constant τ represents the thermal diffusion time in the steady state. For times short compared to the diffusion time, it may represent the laser pulse width. Thus thermal contributions can cause an index change even for short times before a steady state is reached. This is because this index change is proportional to the total energy deposited in the medium (i.e., the integral of the intensity) and increases with time. The quantity $\alpha\tau I$ represents the energy per unit volume absorbed in the material and converted to heat. Thus the index change is in reality fluence dependent. The specific heat C times the mass density ρ converts this energy absorbed into a temperature change ΔT .

Typical values of $|dn/dT|$ range from 10^{-6} to 10^{-3} K^{-1} . The larger values usually correspond to a liquid crystal near the nematic-to-isotropic transition. For $\alpha \sim 10^2 \text{ m}^{-1}$, $\tau \sim 10^{-4} \text{ s}$, $\rho \sim 10^3 \text{ kg/m}^3$, $C \sim 10^3 \text{ J/kg-K}$, and $dn/dT \sim -10^{-4}$; the nonlinear index intensity coefficient is $n_2^I \sim -10^{-12} \text{ m}^2/\text{W}$ and can thus be substantial.

G. Cascaded Second Order Effects

Second order nonlinear optical processes in noncentrosymmetric media were discussed in Chapter 2. It was noted that when perfect phase matching is not achieved, there will be a flow of power between the pump wave(s) and the newly generated frequency wave(s) as the generated wave(s) propagate periodically in and out of phase with the driving nonlinear polarization. This will impress a phase retardation on the pump wave(s) that is intensity dependent.

Consider second harmonic generation as an example of a quasi-self-induced phase retardation. When there is not perfect phase matching, the second harmonic will beat with the fundamental to generate a wave at the fundamental frequency that is in phase quadrature with the incident fundamental wave. The net result is an additional phase retardation impressed on the fundamental. As the incident fundamental intensity increases, a greater fraction of it is converted into the second harmonic within a coherence length. Thus a larger phase retardation is impressed on the fundamental. Such a process is a quasi-third order process

(i.e., nonlinear index of refraction) due to a cascaded second order effect, with an effective third order susceptibility written as $[\chi^{(3)}(-\omega; \omega, \omega, -\omega)]_{\text{eff}} = \chi^{(2)}(-\omega; 2\omega, -\omega) : \chi^{(2)}(-2\omega; \omega, \omega)$.

In the nondepleted pump regime, the pump intensity is essentially constant as a function of position and always large compared to the second harmonic intensity. Under these conditions, the net phase retardation induced by the cascaded process when the wave has traveled through a crystal of length L is given by

$$\Delta\varphi = -\frac{\Delta k L}{2} \left\{ 1 - \left[1 + \left(\frac{2\kappa\sqrt{I_\omega}}{\Delta k} \right)^2 \right]^{1/2} \right\} \quad (38)$$

where $\Delta k = 2k_\omega - k_{2\omega}$, and

$$\kappa = \frac{2\omega d_{\text{eff}}}{\sqrt{2\varepsilon_0 n_\omega^2 n_{2\omega} c^3}} \quad (39)$$

In terms of the vacuum wave vector, this phase change is equivalent to a refractive index change, i.e., $\Delta\varphi = (\omega/c)\Delta n L$. For small enough pump intensity, or large enough phase mismatch, Eq. (38) can be expanded, and to lowest order the resulting Δn is proportional to I_ω . Thus an effective n_2^I can be defined, and its expression is given in [Table 4](#).

Note that the effective nonlinear index intensity coefficient can be either positive or negative depending on the sign of the phase mismatch. In the literature, the phase mismatch is often defined as the negative of that defined above. However, it will always be true that $(n_2^I)_{\text{eff}}$ for SHG will be positive if $n_\omega > n_{2\omega}$ and negative if $n_\omega < n_{2\omega}$.

This effective nonlinear index coefficient can be substantial, with a typical value of $(n_2^I)_{\text{eff}} \sim 10^{-14} \text{ cm}^2/\text{W}$, and feasibly a factor of 1,000 times larger in realistic materials (i.e., for $d_{\text{eff}} \sim 50 \text{ pm/V}$). However, as the incident intensity increases [i.e., $I_\omega \sim (\Delta k/2\kappa)^2$], the expression in Table 4 is no longer valid. Then the expression for the net phase retardation given by Eq. (38) must be used, and the material is not Kerr-like. In general, when pump depletion cannot be ignored, then the coupled wave equations given in Chapter 2 must be solved numerically to find the net nonlinear phase retardation.

It is also noted that other non-phase matched second order processes, such as sum- and difference-frequency generation, produce nonlinear phase shifts. These processes can be considered as equivalent to a strong pump wave inducing a nonlinear index for a weak probe wave.

H. Photorefractive Effect

The photorefractive effect is the last example of a physical mechanism involved in an intensity induced refractive index change. Unlike the previous examples, the index change is not dependent on the magnitude of the intensity that produces it. Rather, it is a nonlocal change in refractive index due to a spatial variation of intensity. It was first observed in 1966 in electro-optic crystals and was described as “optical damage” [7].

The photorefractive effect is often encountered in a situation where two coherent waves of intensities, I_1 and I_2 , overlap and produce interference fringes. The case discussed here is the degenerate one where the frequencies of both waves are the same. The z -components of their wave vectors are parallel, but the x -components point in opposite directions. The intensity pattern produced in the region of the overlap of these two waves is given by

$$I(x) = I_1 + I_2 + 2\sqrt{I_1 I_2} \cos Kx \quad (40)$$

where $K = 2\pi/\Lambda$, and Λ is the fringe spacing. This intensity pattern is stationary in the photorefractive medium.

The photorefractive medium is modeled as a wide gap semiconductor that is normally transparent at the frequency of the incident radiation. However, the semiconductor contains donor and acceptor impurities (electrons or holes), whose energies lie within the forbidden gap of the semiconductor. It is assumed that $N_D \gg N_A$, where N_D and N_A are the number densities of donor and acceptor species, respectively. A small number of donors are thermally ionized, and in the dark the number of these ionized donors equals N_A . The acceptors maintain charge neutrality, but do not contribute directly to the photorefractive effect.

The neutral donors are photoionized by the incident light and create free carriers. Charge transport ensues due to diffusion. If an external field is applied, there will also be a drift current. The discussion here assumes only diffusion. After diffusing a small distance through the crystal, the free carriers can be trapped by ionized donors. In the steady state, this will occur in the dark regions of the intensity pattern. Thus, a space charge density pattern is established. The formation of this space charge density leads to the buildup of an internal space charge field. This field forms an index grating by the linear electro-optic (Pockels) effect. Thus, the medium must be noncentrosymmetric to be photorefractive.

The change in refractive index due to an applied electric field via the Pockels effect is written as

$$\Delta\left(\frac{1}{n^2}\right)_{ij} = \sum_k r_{ijk} E_k \quad (41)$$

where r_{ijk} are elements of the Pockels tensor, which is related to the second order susceptibility $\chi_{ijk}^{(2)}(-\omega; \omega, 0)$, and the subscripts i, j , and k range over x, y, z . In the photorefractive effect, the applied field is the internal space charge field, which is determined by the intensity interference pattern and physical properties of the medium. An externally applied field can modify the space charge field.

In a simple one-dimensional model of the photorefractive effect, the index of refraction can be considered a scalar quantity. For small modulation depth $2(I_1 I_2)^{1/2}/(I_1 + I_2)$, a closed form solution for the refractive index change can be obtained [8] and is given in Table 4, where r_{eff} is an effective Pockels coefficient depending on the polarization and the crystal orientation. (This coefficient can be determined in an analogous way to d_{eff} , as described in Chapter 2, through the use of direction cosines relating the crystal principal axes to the laboratory axes.)

Several things are noted about the refractive index grating $\Delta n(x)$. First, as mentioned earlier, it depends on the intensity only through the modulation depth. The index modulation thus goes to zero when either I_1 or I_2 is zero, and is a maximum when $I_1 = I_2$ (i.e., maximum fringe contrast).

Second, it is noted that $\Delta n(x)$ is $\pi/2$ radians out of phase with the intensity pattern that produced it. This result, due to the diffusion of charge and subsequent trapping in dark regions, leads to a transfer of energy between the two waves incident on the medium. This has several interesting consequences that can be used in a variety of applications (see Section III).

The phase difference between the index modulation and intensity pattern can be understood by a closer examination of the band transport model. For the sake of discussion, assume that the charge carriers are electrons. Free electrons are photogenerated in the bright fringes of the intensity pattern. This creates a free-charge density gradient and leads to diffusion of the electrons, which are then trapped by ionized donors in the dark fringes at a distance of one-half grating period away. Thus, a charge separation is created yielding a charge density grating (alternating positive and negative charge density) that is in phase with the intensity pattern. The charge separation results in a space charge electric field. In the vicinity of the peaks and valleys of the charge density grating, the charge distribution is symmetric. Hence, at the peaks and valleys the field on either side is of equal strength but pointing in the opposite direction. Consequently, the net field here must be zero. On the other hand, in the vicinity of the zeroes of the space charge grating, the charge distribution is asymmetric. The field on either side of these points is of equal strength and pointing in the same direction, and the net field is thus a maximum. For an adjacent zero at a distance $\Lambda/2$ away, the same condition holds except that the net field points in the opposite direction. Hence, the space charge field also forms in an

alternating positive and negative pattern that is $\pi/2$ radians out of phase with the space charge grating and the intensity fringe pattern.

Finally, the band transport model yields an expression for the space charge field amplitude that can be written as

$$E_{sc} = \left(\frac{1}{E_D} + \frac{1}{E_N} \right)^{-1} \quad (42)$$

where E_D and E_N are called the diffusion field and the saturation field, respectively. Expressions for these are given in Table 4. Consequently, the space charge field is less than the smaller of these two fields.

The names given to these fields originated from the physical processes producing them. When a charge density gradient is established charges diffuse in a direction opposite the gradient, creating a diffusion current and leading to charge separation. This charge separation sets up an electric field that opposes the diffusion and establishes a drift current in the opposite direction. The electric field that yields a steady-state net current of zero is called the diffusion field. The steeper the charge density gradient, the stronger this field must be. Thus $E_D \propto \Lambda^{-1}$, and increases for small grating period and decreases for large grating period.

The saturation field E_N is the maximum possible electric field for a given space charge density. By integration of Poisson's equation it is proportional to $eN_A\Lambda/2\pi$, which is the effective areal space charge density (i.e., it has units of coulombs per unit area). Thus, E_N increases with grating spacing since, for a given peak space charge (volume) density eN_A , the net charge contributing to the field increases with Λ .

As a result of these dependencies on Λ , for large grating spacing (small K), $E_D \ll E_N$ and the space charge field approaches E_D . On the other hand, for small grating spacing (large K), $E_D \gg E_N$ and the space charge field approaches E_N . Consequently, the amplitude of $\Delta n(x)$ has a maximum that is reached for a grating wave vector $K_{\max} = (e^2 N_A / \epsilon k_B T)^{1/2}$, where e is the electron charge, ϵ is the average (dc) dielectric constant, k_B is Boltzman's constant, and T is the absolute temperature. The maximum space charge field amplitude is given by $E_{sc,\max} = (N_A k_B T / 4\epsilon)^{1/2}$.

An externally applied field can modify the strength of the space charge field. When $E_N \ll E_D$ the modification is negligible. However, when $E_N \gg E_D$ and the applied field is large compared to both E_N and E_D , then the space charge field amplitude approaches E_N instead of E_D , as it does in the case with no external field. In general, the application of an external field will also shift the relative phase between the index grating and the intensity grating.

The magnitudes of the diffusion and saturation fields can be substantial, ranging from a few to hundreds of kilovolts per centimeter. (Note that the strength of the saturation field is inversely proportional to the dielectric constant and will

thus be smaller in a high dielectric material.) Consequently, the amplitude of the index modulation due to the photorefractive effect can be quite large compared to the nonlinear index changes described earlier for a given incident intensity. However, the process is in comparison quite slow, ranging from tens of microseconds to tens of seconds to reach a steady state (assuming an incident intensity $\sim 1 \text{ W/cm}^2$), depending on the material. The effect depends on charge separation and requires the absorption of at least one photon to separate a charged pair. Thus, the time required to reach a given space charge density is dependent on the photon flux. The actual response time for a given situation is inversely proportional to the incident intensity. It will also depend on other material and geometry parameters, such as the absorption cross section, the quantum efficiency (fraction of photoexcitations resulting in charge separation), the refractive index, the Pockels coefficient, and the grating spacing [8].

III. RELATED PHENOMENA AND APPLICATIONS

The discussion above illustrates several mechanisms whereby the incident light produces a change in the optical properties of a medium. This section examines the reaction of the medium back on the light and describes many of the electromagnetic phenomena resulting from a nonlinear index of refraction.

A. Optical Kerr Effect

Polarized optical radiation can induce a symmetry axis in an isotropic medium. Thus a birefringence is induced by an intense beam of light. This birefringence can lead to two interesting phenomena, intensity induced linear birefringence and ellipse rotation; the two are collectively referred to as the optical Kerr effect. The Kerr effect refers to a birefringence induced by an applied electric field where the index change is second order in the field strength. In the optical Kerr effect, the field derives from the optical frequency electromagnetic wave.

Induced linear birefringence. Consider an isotropic nonlinear medium wherein a strong pump beam at frequency ω and a weak probe beam at frequency ω' propagate collinearly. Both beams are linearly polarized, but the probe polarization is at a 45° angle with respect to the pump polarization. The strong pump establishes an extraordinary axis in the isotropic medium through the nonlinear index of refraction. The probe, which is assumed to be too weak to produce an index change of its own, thus sees the medium as an intensity dependent retardation plate. The two orthogonal components of the probe field will experience a different phase retardation, so that the output probe at the end of the medium will be, in general, elliptically polarized.

The induced linear birefringence is given by the formula in Table 6. It depends on the quantity $\chi_{xyxy}^{(3)} + \chi_{yyxx}^{(3)}$. Thus a measurement of the retardation can yield this quantity for the medium. The net phase retardation at ω' induced by the intensity at ω is also given in Table 6. An application of this effect is an intensity-controlled fast optical shutter. When the material is between parallel polarizers, extinction of the transmitted probe beam occurs at a pump intensity that leads to a net nonlinear phase retardation of $\Delta\varphi = \pi$. If the response of the medium is sufficiently fast, the time response of the shutter will follow the pump pulse temporal profile.

Ellipse rotation. Consider now a single optical beam incident on an isotropic nonlinear medium. For both linearly and circularly polarized light, the third order nonlinear polarization $\mathbf{P}^{(3)}$ has the same vector character as the applied field $\mathbf{E}^{(\sim)}$. Thus the induced birefringence produces no change in the polarization state of the optical field. However, an elliptically polarized wave induces a nonlinear polarization that mixes the left and right circularly polarized components of the wave. The induced circular birefringence Δn_c is given by the formula in Table 6. Note that this quantity depends only on the tensor component $\chi_{xyyx}^{(3)}$. E_+ and E_- are the complex field amplitudes of the left and right circularly polarized components, respectively, of the elliptically polarized wave. The circular birefringence produces a rotation of the polarization ellipse (ellipse axes) through an angle θ_c , which is also given in Table 6. A measurement of this rotation angle gives a measure of the $\chi_{xyyx}^{(3)}$ tensor component for the medium.

Table 6 Formulas Related to the Optical Kerr Effect (SI units)

Induced linear birefringence

Pump (ω) and probe (ω')

Induced birefringence

$$\begin{aligned}\Delta n(\omega') &= \frac{3(\chi_{xyxy}^{(3)} + \chi_{yyxx}^{(3)})}{n_0} |E(\omega)|^2 \\ &= \frac{3(\chi_{xyxy}^{(3)} + \chi_{yyxx}^{(3)})}{2\varepsilon_0 n_0^2 c} I_\omega\end{aligned}$$

Nonlinear phase retardation

$$\Delta\varphi(\omega') = \frac{3\omega' L}{2\varepsilon_0 n_0^2 c} (\chi_{xyxy}^{(3)} + \chi_{yyxx}^{(3)}) I_\omega$$

Probe transmittance

(through parallel polarizers)

$$I_{\omega'}(L) = I_{\omega'}(0) \cos^2\left[\frac{1}{2} \Delta\varphi(\omega')\right]$$

Induced circular birefringence

Single elliptically polarized beam

Induced birefringence

$$\Delta n_c = -\frac{3\chi_{xyyx}^{(3)}}{n_0} (|E_+|^2 - |E_-|^2)$$

Nonlinear phase retardation

$$\theta_c = \frac{\pi \Delta n_c L}{\lambda}$$

B. Four-Wave Mixing

Four-wave mixing refers to the interaction of four waves in a nonlinear medium via the third order polarization. When the waves have different frequencies, the process can be used to generate new frequencies. However, several interesting effects are observed when all the waves have the same frequency. This is referred to as the degenerate case.

Degenerate four-wave mixing in centrosymmetric media. Degenerate four-wave mixing (DFWM) in isotropic media having a center of symmetry is nice to study because it avoids the complications of birefringence and the competing effects of second order processes. Two geometries used in studying this phenomenon are illustrated in Fig. 7. The first is called the backward geometry, since two of the waves travel in the backward direction and two travel in the forward direction. In the second case, all four waves travel in the forward direction, hence its name. This latter case is useful only for thin samples, as will be shown, since it is not phase matched.

The backward geometry is considered first. Two counterpropagating strong pump beams are labeled as the forward pump and the backward pump as shown in Fig. 7a. A third wave called the probe beam is incident at some angle θ to the direction of the forward pump. A fourth beam, called the conjugate beam, is generated in the process and propagates counter to the probe beam.

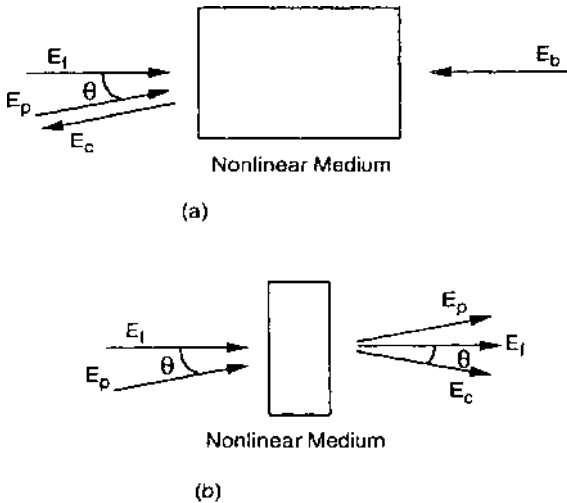


Figure 7 Two configurations for observing degenerate four-wave mixing. (a) Backward geometry. (b) Forward geometry.

The interaction of these waves will generate a third order polarization oscillating at the same frequency ω given by

$$P_c^{(3)} = 6\epsilon_0\chi_{\text{eff}}^{(3)}A_fA_bA_p^*\exp[i(\mathbf{k}_f + \mathbf{k}_b - \mathbf{k}_p)\cdot\mathbf{r}] \quad (43)$$

where

$$\chi_{\text{eff}}^{(3)} = \hat{e}_c \cdot \chi^{(3)}(-\omega; \omega, \omega, -\omega) : \hat{e}_f \hat{e}_b \hat{e}_p^* \quad (44)$$

The polarization is proportional to the conjugate (negative frequency component) of the probe wave so that the net frequency is ω . Note that the degeneracy factor is 6 even though two of the incident waves have the same frequency since they are two distinct fields, traveling in opposite directions. Note that because $\mathbf{k}_f = -\mathbf{k}_b$ and $\mathbf{k}_p = -\mathbf{k}_c$, the DFWM process is automatically phase matched.

Similar third order polarizations can be written for the other three waves. Because of permutation symmetry for this nonresonant process, they will all have the same $\chi_{\text{eff}}^{(3)}$.

The third order polarization couples the four waves. Substituting the expressions for the nonlinear polarization into the wave equation and employing the slowly varying amplitude approximation yields four first order coupled differential equations that describe the spatial evolution of the waves in this process. The usual case involving strong pump beams of approximately equal amplitude allows one to ignore the pump beam equations. This is the nondepleted pump regime where A_f and A_b are approximately constant throughout the medium. The equations for the probe and conjugate waves are then given by

$$\frac{dA_p}{dz} = i\kappa A_c^* \quad (45)$$

and

$$\frac{dA_c^*}{dz} = i\kappa^* A_p \quad (46)$$

where

$$\kappa = \frac{3\omega}{n_0 c} \chi_{\text{eff}}^{(3)} A_f A_b \quad (47)$$

is called the coupling coefficient. In deriving these equations, it is assumed that the angle θ between the probe and forward pump beams is small. Otherwise, the left-hand sides of Eqs. (44) and (45) should be multiplied by $\cos \theta$.

The usual boundary conditions are that the input wave amplitudes are $A_p(0)$ nonzero and $A_c(L) = 0$. Using these conditions, the solutions to the coupled wave

equations are given by

$$A_p(z) = \frac{\cos[|\kappa|(z - L)]}{\cos(|\kappa|L)} A_p(0) \quad (48)$$

$$A_c(z) = -i \frac{\kappa \sin[|\kappa|(z - L)]}{|\kappa| \cos(|\kappa|L)} A_p^*(0) \quad (49)$$

Note that the output wave $A_p(L)$ is amplified by a factor of $1/\cos(|\kappa|L)$. However, the real interest is in the conjugate wave whose output $A_c(0)$ is proportional to the complex conjugate of the incident probe wave. This conjugate wave has been called the time reversed probe wave. It travels in the opposite direction of the probe wave and has exactly the time-reversed phase. It is often referred to as the “reflected” conjugate wave, and the medium in which DFWM occurs is called a “phase conjugate mirror.”

A phase conjugate mirror has several unique properties compared to an ordinary mirror. For example, it functions as a retroreflector for any incident angle. Also, for any angle, if the incident wave is converging (spherical phase curvature), then the reflected wave is diverging along the same path. Any phase distortion $\varphi(\mathbf{r})$ impressed on the incident probe, for example by propagating through an inhomogeneous medium (e.g., turbulence), will result in exactly the opposite phase $-\varphi(\mathbf{r})$ impressed on the reflected wave traveling backward through the distorting medium. Thus the wave emerging from the inhomogeneous medium will have no phase distortions. All of these special effects occur because the conjugate phase of the incident probe beam is impressed on the reflected beam. This is why it is called the conjugate wave.

Another interesting facet of the wave reflected from a phase conjugate mirror is that it can have a larger amplitude than the incident probe. This occurs when $\pi/4 < |\kappa|L < \pi/2$, as can be seen from Eq. (49). Thus both the probe and conjugate waves experience gain under these pump conditions. In fact, when $|\kappa|L \rightarrow \pi/2$ the gain $\rightarrow \infty$. This has an analog in electronic circuitry and refers to the situation where an amplifier bursts into oscillation, i.e., there is a finite output for zero input. Actually, the input is quantum fluctuations (noise). Thus one has the interesting phenomenon of a mirrorless oscillator. The energy for amplifying the beams comes from the pump beams. When the amplification is large, the pumps can no longer be considered nondepleted.

It is useful to define a conjugate reflectance by the ratio of the intensities, I_c/I_p . Expressions for this quantity are given in Table 7. Note that $R \rightarrow \infty$ as $|\kappa|L \rightarrow \pi/2$, reflecting once again the oscillation condition. For small values of $|\kappa|L$ it is noted that $R \sim |\kappa|L^2$. This form is useful in measurements of $\chi_{\text{eff}}^{(3)}$, as will be discussed in Chapter 7.

Table 7 Formulas Related to Four-Wave Mixing^a

Process	Phase conjugate reflectance
DFWM in centrosymmetric media	
No absorption	$R = \tan^2 \kappa L$ $R \approx (\kappa L)^2 \quad \kappa L \ll 1$ $\kappa = \frac{3\omega}{n_0 c} \chi_{\text{eff}}^{(3)} A_f A_b \quad (\text{SI})$ $\kappa = \frac{12\pi\omega}{n_0 c} \chi_{\text{eff}}^{(3)} A_f A_b \quad (\text{cgs})$
Effect of background linear absorption	$R = \frac{4 \kappa L ^2 e^{-\alpha L} \tan^2 \kappa_{\text{eff}} L}{ \alpha L \tan \kappa_{\text{eff}} L + 2\kappa_{\text{eff}} L ^2}$ $\kappa_{\text{eff}} = \sqrt{ \kappa ^2 e^{-\alpha L} - (\alpha/2)^2}$ $R \approx \kappa L_{\text{eff}} ^2 e^{-\alpha L} \quad \kappa L ^2 e^{-\alpha L} \ll (\alpha L/2)^2$ $L_{\text{eff}} = \frac{1}{1 - e^{-\alpha L}}$
Effect of unequal pump beams	$R = \frac{\alpha \tan^2 \kappa' L}{1 + \eta^2 \sec^2 \kappa' L}$ $\kappa' = \kappa \sqrt{1 + \eta^2}$ $\eta = \frac{I_f - I_b}{4\sqrt{I_f I_b}}$
DFWM in photorefractive media (ignoring absorption) (θ = angle between $\mathbf{k}_f$ and $\mathbf{k}_p$)	$R = \frac{\sinh^2 \frac{1}{4} \gamma L}{\cosh^2 [\frac{1}{4} \gamma L - \ln(I_b/I_f)^{1/2}]}$ $\gamma = \frac{\pi}{n_0 \epsilon_0 \lambda \cos \theta} \hat{\mathbf{e}}_p \cdot \boldsymbol{\epsilon}_1 \cdot \hat{\mathbf{e}}_f \quad (\text{see Section III.c})$
DFWM in two-level media	$R = \left \frac{\kappa \sinh \gamma L}{\gamma \cos \gamma L + \alpha \sin \gamma L} \right ^2$ $\gamma = \sqrt{ \kappa ^2 - \alpha^2}$ $\kappa = \alpha_{00} \frac{T_2 \Delta - i}{1 + (T_2 \Delta)^2} \frac{2(I_f I_b)^{1/2} / I_s^\Delta}{\{[1 + (I_f + I_b)/I_s^\Delta]^2 - 4I_f I_b / (I_s^\Delta)^2\}^{3/2}}$ $R = \left \frac{(\kappa/\gamma) \tan \gamma L}{1 + (\alpha/\gamma) \tan \gamma L} \right ^2 \quad \kappa ^2 > \alpha^2$ $R = \left \frac{(\kappa/\gamma) \tanh \gamma L}{1 + (\alpha/\gamma) \tanh \gamma L} \right ^2 \quad \kappa ^2 < \alpha^2$
Nearly DFWM in centrosymmetric media	$R(\delta\omega/\omega) = \frac{\tan^2 \kappa' L}{1 + x^2 \sec^2 \kappa' L}$ $\kappa' = \kappa \sqrt{1 + x^2}$ $x = \frac{k}{ \kappa } \frac{\delta\omega}{\omega}$
Non-DFWM in centrosymmetric media	
Phase conjugate reflectance (Fig. 15)	$R(\omega_1) = \frac{\lambda_2 n_2}{\lambda_1 n_1} \tan^2 \kappa L$

(continued)

Table 7 Continued

Process	Phase conjugate reflectance
Signal amplification (s = signal, p = pump) (Fig. 14)	$\kappa = 3 \left(\frac{\omega_1 \omega_2}{n_1 n_2 c^2} \right)^{1/2} \chi_{\text{eff}}^{(3)} A_3^* A_4$ $I_s(L) = I_s(0) \exp[2\text{Re}(G)]$ $G(L) = i \frac{3\omega_s}{n_s c} \chi_{\text{eff}}^{(3)} A_{p1} A_{p2} L e^{i(\Delta\kappa \hat{z})L/2} \frac{\sin[(\Delta\mathbf{k} \cdot \hat{z})L/2]}{(\Delta\mathbf{k} \cdot \hat{z})L/2}$ $\text{Re}(G) = -\frac{3\omega_s}{n_s c} \text{Im}(\chi_{\text{eff}}^{(3)}) A_p ^2 L, (\Delta\mathbf{k} \cdot \hat{z})L \ll 1$ $= -\frac{3\omega_s \text{Im}(\chi_{\text{eff}}^{(3)})}{2\varepsilon_0 n_p n_s c^2} I_p L \quad A_{p1} = A_{p2} \equiv A_p $

^a Parameters defined in the text. DFWM, degenerate four-wave mixing. (Nondepleted pump approximation.)

Effects of linear nonresonant absorption. If there is some background linear absorption in the medium, the above equations must be modified. This could be due to a nonresonant interaction where the frequency is in the far wings of the absorption line, or it could be due to impurity absorption. For a linear absorption coefficient α , the resulting expression for the conjugate reflectance is given in Table 7. The approximate expression corresponds to the condition $|\kappa L|^2 \exp(-\alpha L) \ll (-\alpha L/2)^2$ and is given in terms of an effective length $L_{\text{eff}} < L$, which is determined by the absorption coefficient: $L_{\text{eff}} = [1 - \exp(-\alpha L)]/\alpha$.

Real-time holography and polarization effects. Some aspects of DFWM can be interpreted as real-time holography [3,9]. This can be observed by rewriting the third order polarization driving the conjugate beam, making use of the symmetry properties of $\chi_{ijkl}^{(3)}$ in an isotropic medium:

$$\mathbf{P}_c^{(3)} = 6\varepsilon_0 [\chi_{xyxy}^{(3)} (\mathbf{E}_f \cdot \mathbf{E}_p^*) \mathbf{E}_b + \chi_{xyyx}^{(3)} (\mathbf{E}_b \cdot \mathbf{E}_p^*) \mathbf{E}_f + \chi_{xyxx}^{(3)} (\mathbf{E}_f \cdot \mathbf{E}_b) \mathbf{E}_p^*] \quad (50)$$

Thus the conjugate wave is due to three independent contributions represented by the three terms in Eq. (50). Note that any one of these contributions can be observed in isolation by appropriately choosing the polarizations of the three incident waves. The first term is selected by choosing $\hat{e}_f \parallel \hat{e}_p \perp \hat{e}_b$. The conjugate wave is then polarized parallel to $\hat{e}_b$. Similarly, the second and third terms are isolated by the choices $\hat{e}_b \parallel \hat{e}_p \perp \hat{e}_f$ and $\hat{e}_f \parallel \hat{e}_b \perp \hat{e}_p$, respectively. In each case, the conjugate wave assumes the polarization of the last of the unit vector triplets.

The first term of Eq. (50) is interpreted as the scattering of the backward pump beam from a Bragg grating written by the interference of the forward pump and probe beams, where the phase grating is recorded through the nonlinear index of refraction. Similarly, the second term corresponds to a Bragg grating written by the interference pattern due to the overlap of the backward pump and probe

beams, which then scatters the forward pump beam. In both cases these are time independent stationary gratings in space, the first having a coarse grating spacing and the second having a fine grating spacing. These are illustrated in Fig. 8. The scattered pump beams are always counter propagating to the incident probe beam because of the angular selectivity of the Bragg gratings and the phase matching of the interaction.

This real-time grating picture also helps explain the phase conjugation of the reflected beam. This is illustrated in Fig. 9. If the phase of the probe beam is distorted, this phase distortion is imprinted in the phase gratings recorded by the interference of the probe with both pump beams. Each pump beam acts as a reading beam for this hologram. Since each pump beam reads from the opposite direction of the other pump, its scattered component carries the phase information present in the hologram, but in an exactly opposite sense to that of the incident probe, as illustrated in Fig. 9.

The last term in Eq. (50) does not bear any analogy to holography. There are two ways in which it can be interpreted, both of which are valid depending on the type of medium in which the interaction takes place. In its present form, written in Eq. (50) it is apparent that the forward and backward waves will interfere to form a grating stationary in space. However, since the conjugate of either wave is not involved, this grating is not time independent but rather oscillates at 2ω . Secondly, the probe wave is not incident at the correct angle for Bragg scattering into 180° . Therefore at first glance a grating interpretation does not appear to be appropriate.

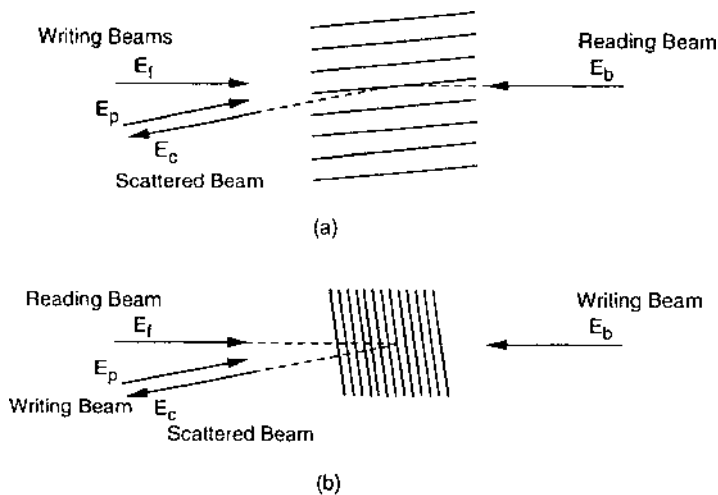


Figure 8 Real-time holographic gratings recorded in a nonlinear medium by degenerate four-wave mixing. (a) Coarse grating. (b) Fine grating.

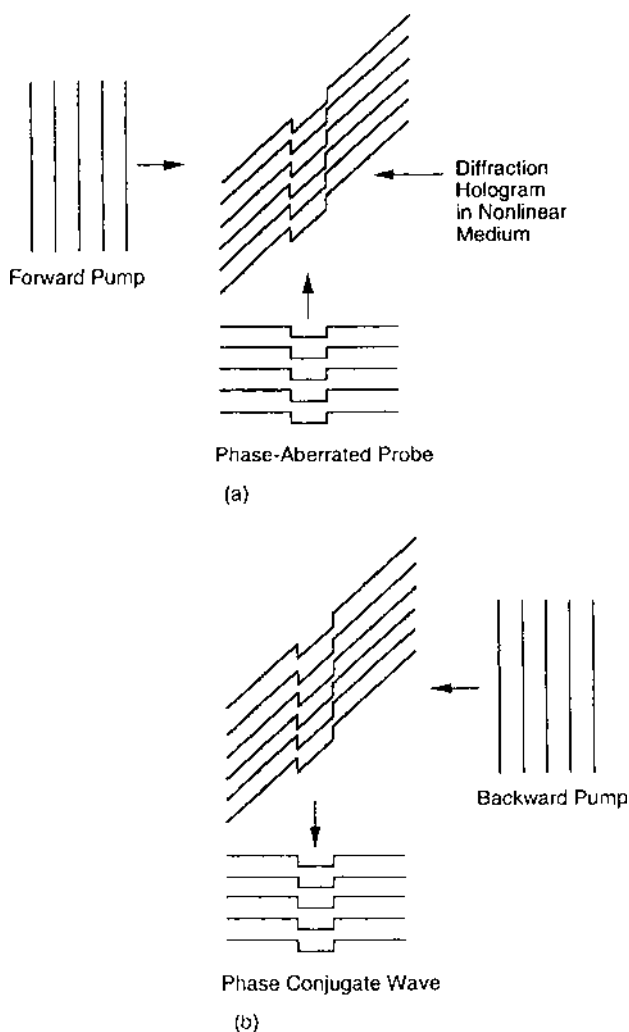


Figure 9 Grating formation and readout (phase conjugate wave) for a phase-distorted probe wave. (a) Grating recording. (b) Grating readout.

Rather the phase conjugate beam is due to a scattering of the probe from a two-photon excitation [10]. This term will thus be strong when the material has a nearby two-photon absorption resonance. Quantum mechanical considerations indicate that if the two-photon resonance is strong, then the material will be put into a two-photon coherent state (i.e., a superposition of the ground and

two-photon excited states). This coherent state will oscillate at 2ω . The incident probe photon stimulates a scattering of two photons from this state: one in the forward probe direction and one in the backward probe direction. The most efficient scattering takes place along the direction that is phase matched and along the direction that will achieve gain (i.e., along the probe beam axis).

Even when a two-photon resonance is not nearby, the last term in Eq. (50) can make a significant contribution to the conjugate wave. To see this, first rewrite this term as $\chi_{xyx}^{(3)} \mathbf{E}_f \cdot (\mathbf{E}_b \mathbf{E}_p^*)$. This now has the appearance of the forward beam scattered from a tensor grating formed by the backward pump and probe beams. This grating is again independent of time and stationary in space. However, it is fundamentally different from those described earlier. Since the backward pump and probe wave are assumed to be polarized orthogonally, their superposition will not produce interference fringes. They will, however, produce a field vector whose direction varies periodically along the phase front of the forward pump wave. A medium of anisotropic molecules will attempt to reorient with this rocking polarization and thus produce a periodically modulated birefringence that scatters the forward beam in the direction opposite to the probe propagation. Likewise, the forward pump and probe beams will also form such a grating to scatter the backward pump. These types of gratings are called orientation gratings [11,12]. A medium with anisotropic absorption will also respond to these vector patterns and produce strong orientation gratings.

Effects of two-photon absorption. This grating picture takes on more interesting aspects when strong two-photon absorption is present. It should be noted that the conjugate reflectance is related only to the modulus of the third order susceptibility through the factor $|\kappa|$. The third order susceptibility is complex, in general. Its real part relates to the nonlinear index coefficient discussed above. The imaginary part gives rise to two-photon absorption (see Chapter 9). Thus the diffraction gratings referred to earlier are made up of two contributions that are in phase quadrature (i.e., $\pi/2$ radians out of phase). Thus they contribute independently in a time averaged sense. The refractive index part creates a phase grating. The two-photon absorption part creates what is known as an amplitude grating that contributes to Bragg scattering but also produces optical loss.

It is possible to observe this amplitude grating in solutions using picosecond pulses. In a solution, the conjugate signal is due to nonlinearities contributed by both solvent and solute molecules. Usually, the solvent is chosen to have negligible two-photon absorption. If the real parts of the solvent and solute contributions to $\chi^{(3)}$ have opposite signs, then for some concentration of solute, the two real parts will cancel. The detected conjugate signal then comes from scattering off the amplitude grating due to the imaginary part of $\chi^{(3)}$. To see this effect, the conjugate signal must be measured as a function of concentration [2].

Strong two-photon absorption can lead to other changes in the linear optical properties of the medium. This cascading of effects of different orders creates an effective fifth order process in the medium. One example is the creation of a thermal grating due to the heating of the medium caused by the two-photon absorption amplitude grating. As discussed in Section II.F, the resultant temperature distribution will produce an index distribution through the thermo-optic effect. Another possibility is a concentration grating of new species with a different linear susceptibility than that of the surrounding medium. This could be a two-photon excited state, a triplet state of the molecule, or possibly a different conformation of the molecule.

The index amplitude corresponding to the phase grating created by this effective fifth order effect can have the opposite sign of that due to the real part of $\chi^{(3)}$, i.e., the two phase gratings can be π radians out of phase. When this is the case, at some pump intensity the two gratings will have the same strength and thus wash each other out. All that is left is the two-photon amplitude grating to scatter the incoming pump beams. When the DFWM process is created with optical pulses, this washout will not be instantaneous, since the fifth order phase grating takes some time to build up. The growth of this grating can be observed using nanosecond pulses. Depending on the pulse width and the energy in the pulses, it is possible to isolate the amplitude grating. The effect is easy to see by capturing and digitizing the phase conjugate pulse. The pulse exhibits a double peak, where the dip corresponds to the point in time when the phase gratings disappear [13]. This point will move to earlier times in the pulse as the pump pulse energy is increased. Examples are shown in Fig. 10.

Effects of unbalanced pump beams. All of the effects described above assumed equal pump amplitudes. When this is not the case, the conjugate reflectance is affected even for nondepleted pumps. The expression for the conjugate reflectance under these conditions is given in Table 7.

To be complete, the nonlinear polarizations leading to self-action and cross-action of the beams (i.e., optical Kerr effect) should be included in the coupled wave equations. These were ignored in the treatment above. The self-action of each beam just contributes a constant phase shift to the waves and hence does not create any observable effect to the DFWM process, unless polarization effects are detected. Hence these can safely be ignored. The combined effects of self-action and cross-action cannot be ignored when the pump beams are unequal.

This combination produces a z dependent phase shift, proportional to the asymmetry in the pump beam intensities, which appears in the product of the pump amplitudes. This adds a phase term to the coupling coefficient κ , which has the effect of introducing a phase mismatch between the driving nonlinear polarization and the phase conjugate wave. Hence this produces a decrease in the efficiency of the conjugate wave production. This intensity dependent phase

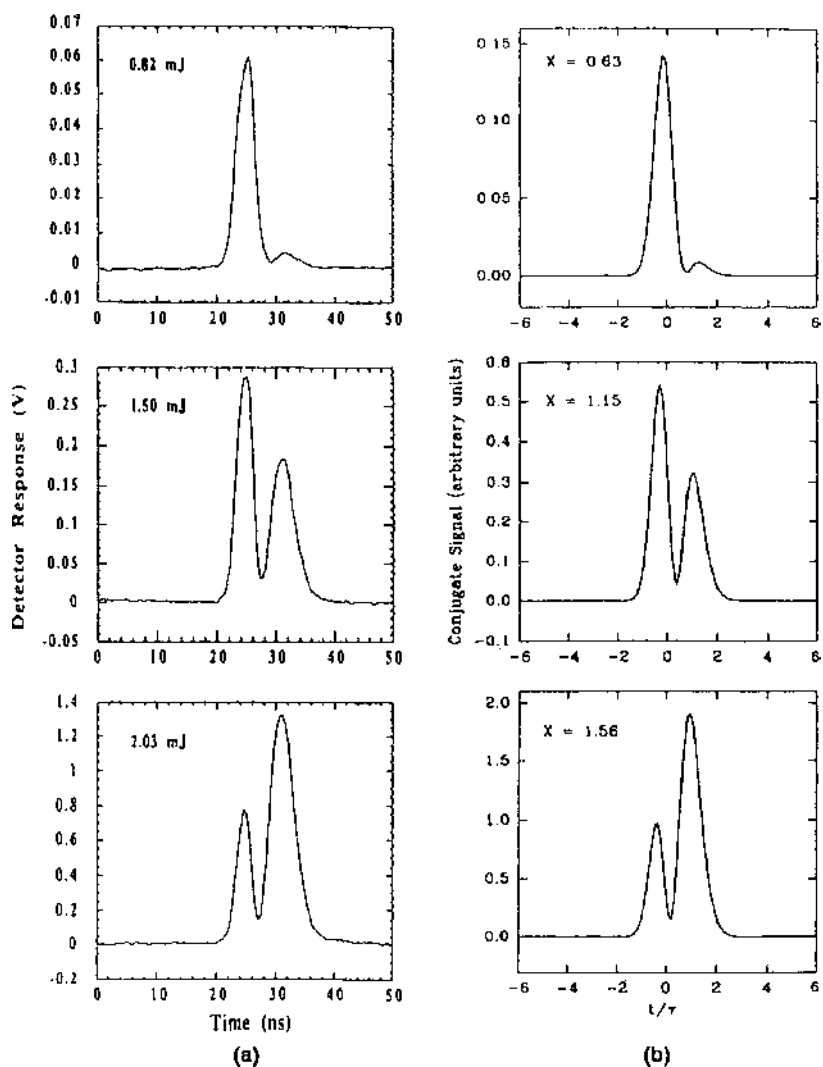


Figure 10 Effects of two-photon absorption on pulsed degenerate four-wave mixing in the nanosecond regime (solution of diphenyl butadiene in chloroform). The left set of plots are experimental data showing the effects of increasing pump energy, and the right set are theoretical calculations. The parameter X is proportional to pump energy, and both X and pump energy are in the ratio of 1:1.83:2.48 (From Ref. 13.)

mismatch can be offset by an intentionally introduced linear phase mismatch. This could be, for example, a small angle adjustment of the pump beams from exact counterpropagation, or a small frequency shift in the probe wave.

Pump depletion [14]. The four coupled-wave equations cannot be solved analytically when pump depletion is included. Numerical analyses predict that it is possible to fully convert the power of one of the pump waves into the conjugate wave. However, full conversion is only possible if the two pump intensities are equal and if the incident probe intensity is small compared to $[(4/3)\epsilon_0 n_0 c I_f(0)/\chi_{xxxx}^{(3)}]^{1/2}$. Oscillation is still possible but will be suppressed for a nonzero input probe intensity. For increasing pump intensity, power will flow back and forth from the pump waves to the probe and conjugate waves.

DFWM in photorefractive media [8]. The sinusoidal index modulation recorded in a photorefractive medium by two interfering waves was described in Section II.H. In the notation of backward DFWM, these recording waves correspond to the forward pump and probe beams. This coarse phase grating (a “transmission grating”) scatters a beam propagating counter to the forward pump beam into the direction of the phase conjugate beam in the backward DFWM geometry. Simultaneously, the backward wave records a fine grating (a “reflection grating”) by interfering with the probe beam, which then scatters the forward pump in the direction of the conjugate beam. In many practical situations, this grating contributes negligibly to the phase conjugate signal since, in the limit of small K , the space charge field responsible for the index modulation is small in comparison to that associated with the coarse grating.

The index modulation is due to the photorefractive effect. This is a real stationary spatial modulation. Hence, there is no two-photon coherence or orientation grating contributing to the DFWM process in photorefractive media. There are, however, some unique aspects of the photorefractive effect that also make DFWM different in these media.

As noted in Section II.H, the index modulation is shifted in phase by $\pi/2$ radians from the intensity pattern that recorded it. Section III.C describes how this leads to a coupling between the two recording beams. This coupling produces a one-way power flow from one recording beam to the other. Thus, the conjugate wave is due not only to the scattering of a third wave incident on the medium, like that described above for Kerr media, but also due to this self-scattering process called two-beam coupling.

The expression for the phase conjugate reflectance due to DFWM in photorefractive media in the nondepleted pump approximation, including the effects of unequal pump intensities, is given in Table 7. Only contributions from the coarse grating have been included. Note that for symmetric pumping, the reflectance is always less than one. Larger reflectances are obtained using asymmetric pumping. Still, the reflectance cannot be driven to ∞ where oscillation occurs. To achieve this condition, a phase shift other than $\pm \pi/2$

between the phase grating and the intensity modulation, as well as unequal pump intensities, must be obtained. Such a phase shift can be achieved by applying an external field or making the frequencies of the interacting waves nondegenerate.

Some of the differences between DFWM in Kerr media and photorefractive media bear repeating. In Kerr media, the phase conjugate reflectance increases with pump intensity. This is not so in photorefractive media since the coupling coefficient is independent of intensity. For a 1:1 pump beam ratio the reflectance in Kerr media can go to ∞ , producing oscillation. The oscillation condition is defined by a critical index modulation (coupling coefficient) achieved at some finite pump intensity. In a photorefractive medium the reflectance never exceeds unity for this pump ratio. Whereas a pump beam imbalance is detrimental to the reflectance in Kerr media, it can produce a reflectance $\gg 1$ in photorefractive media. The condition for maximum reflectance is $\gamma L = 2 \ln(I_b/I_f)$. This condition is also required for self-oscillation. In addition, oscillation requires a particular value of not only the index modulation (which in this case is independent of pump intensity), but also a particular phase difference unequal to $\pm \pi/2$ between the phase grating and the intensity modulation.

The gratings in photorefractive DFWM are actually tensor gratings, determined by the polarizations of the recording beams and the orientation of the crystal. The conjugate reflectance as well as its polarization will depend on these factors. Additional aspects of wave mixing in photorefractive media are quite extensive, and the reader is referred to the literature for further details [8].

Resonant saturation case. The nonlinear refractive index of a two-level system was taken up in section II.E. Only the case of homogeneous absorption line broadening is considered here.

Both amplitude and index gratings are recorded near resonance. The conjugate reflectance is given in Table 7. The effective coupling coefficient is related to the saturable index and the saturable absorption coefficient.

The conjugate reflectance does not necessarily increase without limit in this case because of saturation. When $|\kappa|^2 > (\alpha/2)^2$, then R can become infinite when $\tan \gamma L = -2\gamma/\alpha$, where $\gamma^2 = |\kappa|^2 - (\alpha/2)^2$, κ is the complex coupling coefficient, and α is the saturable absorption coefficient. (Expressions for these quantities are given in Table 7.) When $|\kappa|^2 < (\alpha/2)^2$, R cannot become infinite for any value of γL [15].

A pure amplitude grating is possible at resonance ($\Delta = 0$). The general behavior of the reflectance for such gratings is shown in Fig. 11. For $I > I_s^\Delta$, the reflectance decreases, since the absorption coefficient, acting like the coupling coefficient in this case, saturates and becomes smaller. Also, as $\alpha_0(0)L$ increases, where $\alpha_0(0) \propto N$ (number density) is the on-resonance low-intensity absorption coefficient, the reflectance increases due to an increase in N (i.e., more absorbers). However, this is eventually offset by increased absorption of the probe

and conjugate waves, so increasing $\alpha_0(0)L$ gives diminishing returns. Here the reflectance can never exceed unity.

The reflectance can only exceed unity by detuning from line center, if this is done while also increasing the density. Figure 11 shows this effect by plotting the reflectance as a function of I/I_s^Δ for different values of $\alpha_0(0)L/(1 + T_2^2\Delta^2)$. The cost of increased reflectance is a higher pump intensity requirement, since the saturation intensity scales as $1 + T_2^2\Delta^2$.

The above discussion ignores the absorption of the pump beams. It turns out that this is significant for $I/I_s^\Delta < 1$. At higher intensities, saturation makes pump losses insignificant.

Motion of the atoms in a gas can have a significant effect on the conjugate reflectance. This is due to two effects. One is that Doppler broadening (inhomogeneous broadening) causes the absorption to saturate as $(1 + I/I_s^\Delta)^{-1/2}$ rather than $(1 + I/I_s^\Delta)^{-1}$. The other reason is that the motion tends to wash out the fine grating. Details of these processes can be found in the literature [11]. The effects of equal beam intensities and pump depletion are also discussed in Ref. 11.

Forward conjugate wave generation. Forward DFWM was discussed briefly above. It is illustrated again in more detail in Fig. 12. Forward DFWM involves only a single pump beam, the forward pump shown in the figure. The probe beam is incident on the medium from the same side and overlaps the forward pump. The output waves include, in addition to the transmitted pump and probe, a conjugate wave and a fourth wave called the auxiliary wave. In actuality, for an optically thin medium, there can be a multitude of output waves. The pump and probe waves write an index grating in the nonlinear medium that self-diffracts these waves. The scattering from this optically thin nonlinear grating corresponds to Raman–Nath diffraction, which produces a multitude of diffracted waves. For most practical situations, consideration of the waves depicted in Fig. 12 is sufficient.

The usual situation is that of a strong pump and a weak probe. Thus $|E_f| \gg |E_p|$, which implies that $|E_f| \gg |E_c|$ and $|E_c| \gg |E_a|$ also [5]. Under these conditions, the pump can be considered nondepleted, and only wave equations for E_f , E_p , and E_c need to be solved. The complex wave amplitudes are determined to be

$$A_f(z) = |A_f(0)|\exp(ikz) \quad (51)$$

$$A_p(z) = A_p(0)\exp\left[i\left(\kappa + \frac{\Delta k}{2}\right)z\right]\left[\cosh(gz) + i\frac{(\kappa - \Delta k/2)}{g}\sinh(gz)\right] \quad (52)$$

$$A_c(z) = i\frac{\kappa}{g}A_2^*(0)\exp[i(\kappa + \Delta k/2)z]\sinh(gz) \quad (53)$$

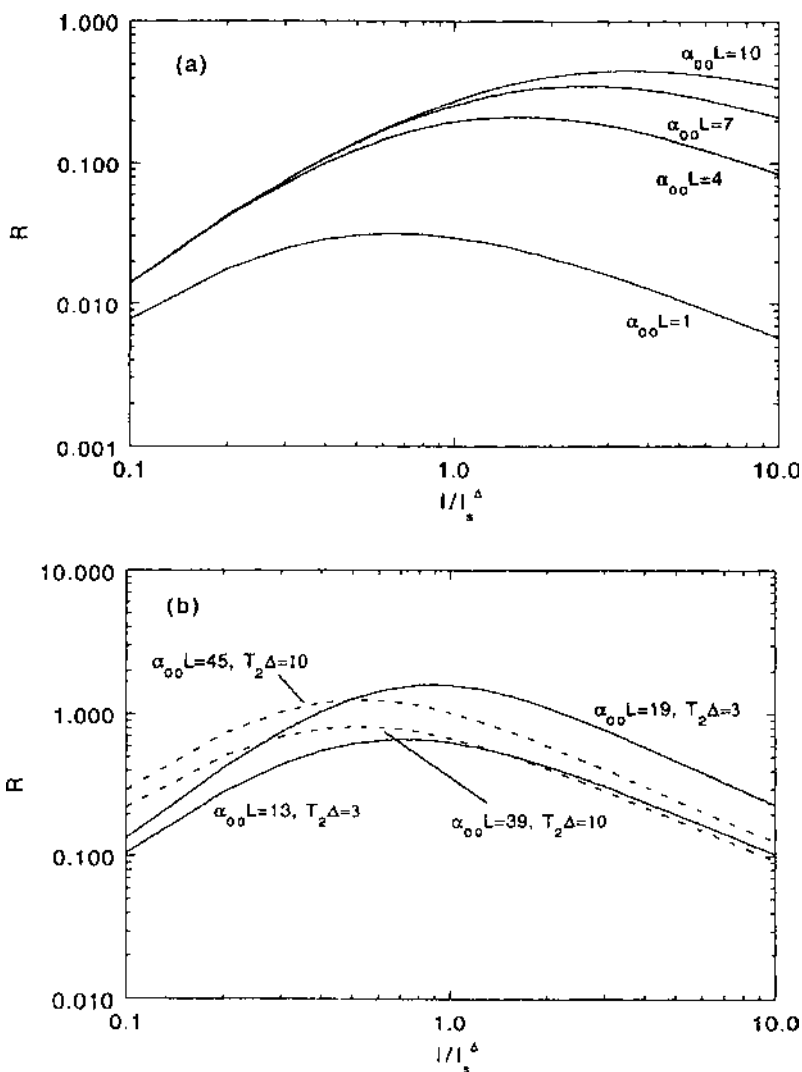


Figure 11 Phase conjugate reflectance in a homogeneously broadened two-level system as a function of intensity. (a) Pure amplitude gratings ($T_2\Delta = 0$). (b) Mixed amplitude and phase gratings ($T_2\Delta \neq 0$).

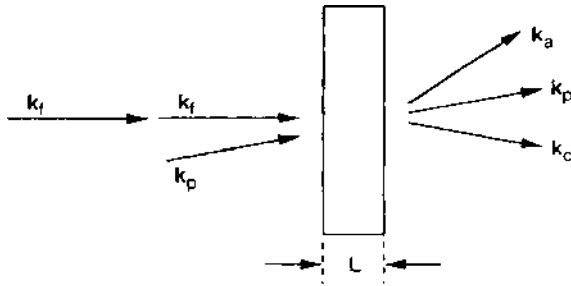


Figure 12 Forward degenerate four-wave mixing in a thin nonlinear medium.

where

$$\kappa = \frac{3\omega}{n_0 c} \chi_{\text{eff}}^{(3)} |A_f(0)|^2 \quad (54)$$

is the nonlinear coupling coefficient,

$$\Delta k = (2\mathbf{k}_f - \mathbf{k}_p - \mathbf{k}_c) \cdot \hat{z} \quad (55)$$

is the phase mismatch, and

$$g = \sqrt{\Delta k \left(\kappa - \frac{\Delta k}{4} \right)} \quad (56)$$

Notice that all of the waves experience a nonlinear phase shift due to the strong pump beam. Also, the conjugate wave is proportional to the complex conjugate of the input probe wave. If the medium is thin, so that $\Delta k L$ is small, this wave can be appreciable. The probe wave is amplified by the interaction with a gain factor G given by

$$G = \frac{I_p(L)}{I_p(0)} = 1 + \frac{\kappa^2}{g^2} [\cosh^2(gL) - 1] \quad (57)$$

Nearly degenerate four-wave mixing. In this case, the pump waves have frequency ω , while the incident probe wave is taken to have a frequency $\omega - \delta\omega$, where $\delta\omega/\omega \ll 1$. This introduces a small phase mismatch to the interaction, which profoundly affects the conjugate reflectance. The corresponding formula is given in Table 7. This is shown plotted in Fig. 13 as a function of $k\delta\omega/\omega|\kappa|$. It can be seen that now the conjugate mirror acts as narrow band reflector, with a peak at

$\delta\omega = 0$ and a bandwidth of

$$(\delta\omega)_{\text{FWHM}} = \frac{2c|\kappa|}{n_0} \left[1 + R_{\text{peak}} - (2 + R_{\text{peak}}) \frac{\tan^{-1} \sqrt{R_{\text{peak}}}}{\sqrt{R_{\text{peak}}}} \right]^{-1/2} \quad (58)$$

for $\delta\omega/\omega \ll 1$, where $R_{\text{peak}} = \tan^2|\kappa L|$.

Nondegenerate four-wave mixing. For completely nondegenerate frequencies, other geometries must be considered to obtain efficient wave mixing. Two examples are considered here.

The first is illustrated in Fig. 14. Here two pump beams of frequency ω_p and a signal beam of frequency ω_s are incident on an isotropic medium. The third order susceptibility that couples the waves and gives rise to an output signal wave $E_s(L)$ is $\chi_{ijkl}^{(3)}(-\omega_s; \omega_s, \omega_p, -\omega_p)$. The pump waves are designated E_{p1} and E_{p2} since, although they have the same frequency, they may be distinct fields having unique directions and polarizations. The phase mismatch is thus $\Delta\mathbf{k} = \mathbf{k}_{p1} - \mathbf{k}_{p2}$.

In the nondepleted pump approximation the output signal field is related to the input signal field by an exponential factor $G(L)$ (see Table 7). Note that this factor is related to a sine function, which accounts for the phase mismatch just as in second order frequency mixing processes. Thus the factor $G(L)$ is effective only over a distance of the order of a coherence length. If the geometry is such that phase matching is achieved, or if the material is thin compared to a coherence

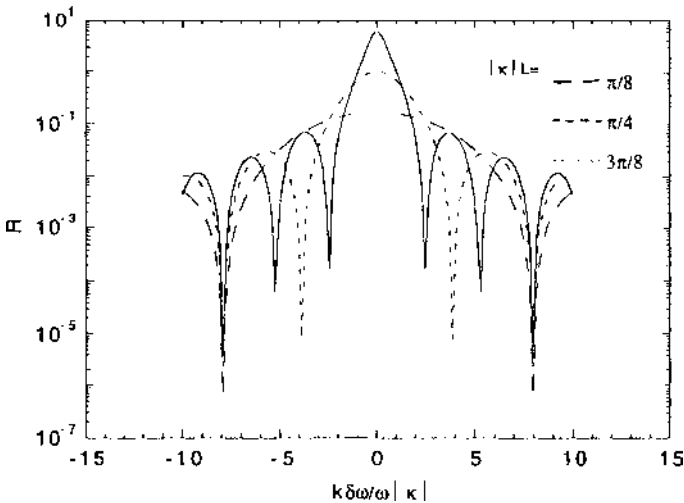


Figure 13 Narrow band phase conjugate reflectance as a function of detuning from exact phase matching for nearly degenerate four-wave mixing.

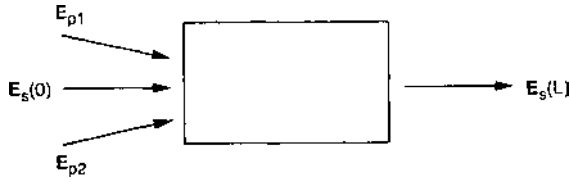


Figure 14 Nondegenerate four-wave mixing geometry for gain or loss processes.

length, then the signal wave will experience gain or loss depending on the sign of $\text{Im}(\chi_{\text{eff}}^{(3)})$, where $\chi_{\text{eff}}^{(3)} = \hat{e}_s \cdot \chi^{(3)} : \hat{e}_s \hat{e}_{p1} \hat{e}_{p2}^*$. A negative imaginary part corresponds to gain, while a positive imaginary part corresponds to loss. A specific example of this is stimulated Raman scattering, for which $\chi_{\text{eff}}^{(3)}$ is resonantly enhanced and phase matching is automatically satisfied (see Chapter 14).

A second nondegenerate four-wave mixing example is shown in Fig. 15, where the input waves are the pump waves E_3 and E_4 and the signal waves $E_1(0)$ traveling to the right and $E_2(L)$ traveling to the left. The frequencies are such that $\omega_4 = \omega_1 + \omega_2 + \omega_3$. Hence the signal waves couple to the pump waves in the combination $E_3^* E_4$ (i.e., $\omega_1 = \omega_4 - \omega_3 - \omega_2$ and $\omega_2 = \omega_4 - \omega_3 - \omega_1$). A typical phase matching geometry for this process is shown in Fig. 15, where the signal waves are counterpropagating. Nondepleted pumps are assumed.

The two signal waves are coupled in the interaction, and the output wave amplitudes are given by

$$A_1(L) = \frac{A_1(0)}{\cos|\kappa L|} + i \frac{\kappa}{|\kappa|} \left(\frac{\omega_1 n_2}{\omega_2 n_1} \right)^{1/2} A_2^*(L) \tan|\kappa L| \tag{59}$$

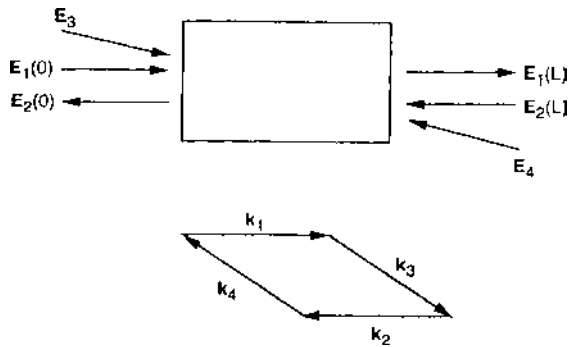


Figure 15 Nondegenerate four-wave mixing geometry for a mirrorless parametric oscillation process.

$$A_2^*(0) = -i \frac{|\kappa|}{\kappa} \left(\frac{\omega_2 n_1}{\omega_1 n_2} \right)^{1/2} A_1(0) \tan|\kappa L| + \frac{A_2^*(L)}{\cos|\kappa L|} \quad (60)$$

where

$$\kappa = 3 \left(\frac{\omega_1 \omega_2}{n_1 n_2 c^2} \right)^{1/2} \chi_{\text{eff}}^{(3)} A_3^* A_4 \quad (61)$$

is the complex coupling coefficient. An interesting case is when $A_2(L) = 0$. Then Eq. (60) predicts that the output field $A_2(0)$ is proportional to the complex conjugate of the input signal wave $A_1(0)$. The conjugate reflectance, given in [Table 7](#), looks formally the same as in the DFWM case, except that it is scaled by the ratio of the wavelengths and refractive indices.

An even more interesting situation occurs when both input signal waves are zero. Note that when the pump intensities are sufficiently high so that $|\kappa L| = \pi/2$, Eqs. (59) and (60) predict infinite gains for the signal waves. This is the oscillation condition once again. This case thus corresponds to a third order mirrorless optical parametric oscillator. The feedback is provided by the counterpropagating coupled waves.

Optical phase conjugation. One of the most studied applications of DFWM is the correction of beam (phase) aberrations by phase conjugation. The properties of the phase conjugate wave that are exploited were described above. The probe beam, which passes through an aberrating medium, is incident on a phase conjugate mirror. The reflected phase conjugate wave passes back through the aberrating medium in the opposite direction. Since the phase is reversed on this wave, the aberrating medium then cleans up the phase aberrations. This is illustrated in [Fig. 16](#).

The assumption of infinite plane waves and nondepleted pump waves gives the impression that the process of phase conjugation is exact. Actually, this is not the case, and several factors limit the quality, or fidelity, of the phase conjugation. Some of these are listed in [Table 8](#). A discussion of these is beyond the scope of this book. They are adequately treated in the literature on optical phase conjugation [16].

The literature on other applications of optical phase conjugation is also extensive. The richness and diversity of these ideas is illustrated in [Table 9](#). It should be noted that in some cases these ideas have been investigated utilizing the phase conjugation attributes of stimulated Brillouin scattering (see Chapter 15) rather than DFWM. However, the ideas are quite general and independent of the choice of nonlinear medium.

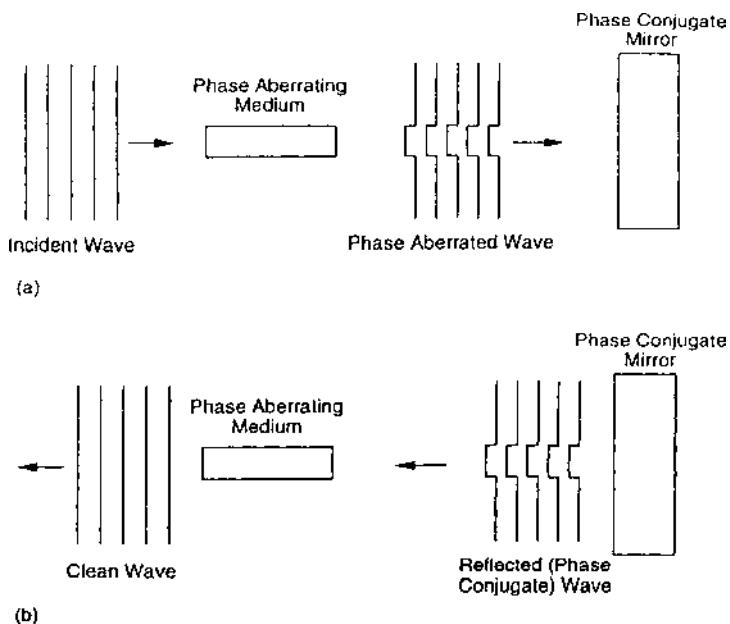


Figure 16 Illustration of aberrated phase cleanup by optical phase conjugation. (a) Phase aberrated wave incident on phase conjugate mirror. (b) Cleaned up phase aberrated wave reflected from phase conjugate mirror.

Table 8 Some Factors Affecting the Quality of Phase-Corrected Beam Aberrations by Optical Phase Conjugation [14]

Factor	Ref.
Limiting aperture (preventing some rays from reaching interaction region)	[62–64]
Nonexact retrace of conjugate beam through aberrating medium	[65]
Motion of aberrating medium	[14]
Nonlinear phase shifts	
Phase mismatch	[66]
Self-focusing/defocusing	[63,67]
Birefringence (ellipse rotation)	[68]
Phase aberration of pump beam(s)	[65,69,70]
Saturation	[63,71,72]

Table 9 Some Applications of Optical Phase Conjugation

Aberration compensation (phase, polarization)
Lensless imaging (high resolution photolithography)
Spatial information processing
Image addition, subtraction, differentiation (edge enhancement), integration
Spatial signal convolution and correlation
Intensity filtering
Logic and digital processing of binary signals
Coherent optical processing (using iterative algorithms)
Frequency filtering
Temporal signal processing
Pulse envelope shaping
Optical gating and time delay control
Pulse envelope time reversal
Temporal signal convolution and correlation
Pulse spreading compensation
Phase conjugate resonators
Photorefractive phase conjugators
Self-pumped
Mutually pumped
Phase conjugate Fabry–Perot interferometer (retroreflected intensity has same form as transmitted intensity for ordinary Fabry–Perot)
Phase conjugate Michelson interferometer (output intensity is always zero, retroreflection is phase conjugate of input; useful for parallel image subtraction)
Phase conjugate Sagnac interferometer (output intensity is zero for 50/50 beam splitter; useful for detecting nonreciprocal phase shift due to rotation of interferometer)
Phase conjugate Mach–Zehnder interferometer (useful for image subtraction)

C. Two-Beam Coupling

This phenomenon involves the interaction of two waves in a nonlinear medium with a nonreciprocal transfer of energy from one wave to the other. Photorefractive and Kerr media are considered.

Photorefractive media. Two beams are incident on a photorefractive crystal as illustrated in Fig. 17. They cross at an angle 2θ in the crystal. As described above in Section II.H, these beams write an index grating in the crystal that is phase shifted from the sinusoidal intensity pattern. The beams self-diffract from this grating by Bragg scattering. One beam is amplified, and the other is attenuated. The direction of energy flow depends on the phase of the grating and the orientation of the crystal.

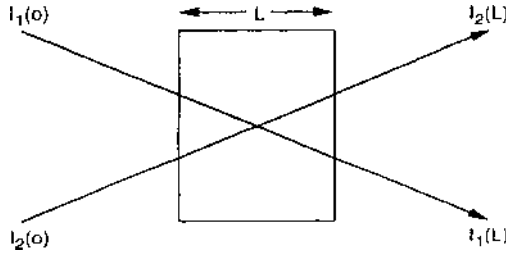


Figure 17 Two-beam coupling in a photorefractive medium.

For the configuration shown in Fig. 17, called codirectional beam coupling, the coupled wave equations are given by

$$\frac{dI_1}{dz} = -\gamma \frac{I_1 I_2}{I_1 + I_2} - \alpha I_1 \quad (62)$$

and

$$\frac{dI_2}{dz} = \gamma \frac{I_1 I_2}{I_1 + I_2} - \alpha I_2 \quad (63)$$

where α is the linear absorption coefficient, assumed to be the same for both beams, and γ is the photorefractive coupling coefficient. In terms of a sinusoidal dielectric grating with amplitude ϵ_1 [8], this coefficient is given by

$$\gamma = \frac{\pi}{n_0 \epsilon_0 \lambda \cos \theta} \hat{e}_1 \cdot \epsilon_1 \cdot \hat{e}_2 \quad (64)$$

where the components of the tensor grating amplitude are given by

$$(\epsilon_1)_{ij} \exp(i\varphi) = -2 \sum_k \epsilon_0 n_i^2 n_j^2 r_{ijk} E_k^{sc} \quad (65)$$

The phase of the grating is φ , which is determined by the space charge field, of which E_k^{sc} is the k th component, and r_{ijk} is the component of the electrooptic tensor (Pockels coefficient). The direction of power flow is determined by the sign of γ , which can be reversed by rotating the crystal through 180° . For a space charge field that is determined by charge diffusion only, the phase is $\varphi = \pi/2$. The expression for the diffusion-controlled space charge field was given in Section II.H. Examples of the coupling coefficient for specific cases for crystals of point group symmetry $4mm$ (e.g., BaTiO_3 or strontium barium niobate [SBN]) and for cubic crystals are given in Table 10.

The solutions of Eqs. (62) and (63) for codirectional beam coupling in a photorefractive medium are given in Table 11 in terms of the input intensity ratio,

Table 10 Form of the Coupling Coefficient γ for Photorefractive Two-Beam Coupling for Two-Crystal Classes

<i>4mm</i> (Plane of incidence = <i>ac</i> plane of crystal)
(1) Incident light polarized $\perp$ plane of incidence $(\alpha = \text{angle between } \mathbf{K} \text{ and } c\text{-axis}) \gamma = -\frac{2\pi}{\lambda \cos \theta} (n^o)^3 r_{13} E^{\text{sc}} \cos \alpha$
(2) Incident light polarized $\parallel$ plane of incidence $(\alpha = \text{angle between } \mathbf{K} \text{ and } c\text{-axis}) \gamma = -\frac{2\pi}{n\lambda \cos \theta} [\cos \theta_1 \cos \theta_2 (n^e)^4 r_{13} \cos \alpha$ $+ (n^o)^2 (n^e)^2 r_{42} \sin \alpha \sin 2\alpha + \sin \theta_1 \sin \theta_2 (n^e)^4 r_{33} \cos \alpha] E^{\text{sc}}$ $\theta_1 = 90^\circ + \theta - \alpha$ $\theta_2 = 90^\circ - \theta - \alpha$
<i>$\bar{4}3m$</i> (Cubic crystals)
$\gamma = \frac{\pi}{\lambda} n^3 r_{41} E^{\text{sc}}$

Table 11 Formulas Related to Two-Beam Coupling in Photorefractive and Kerr Media

<i>Photorefractive media (degenerate codirectional coupling)</i>
$I_1(L) = I_1(0) \frac{1+m^{-1}}{1+m^{-1}e^{\gamma L}} e^{-\alpha L}$
$I_2(L) = I_2(0) \frac{1+m}{1+me^{-\gamma L}} e^{-\alpha L}$
$m = \frac{I_1(0)}{I_2(0)}$
<i>Photorefractive media (degenerate contradirectional coupling)</i>
$I_1(L) = I_1(0) \frac{1+m^{-1}}{1+m^{-1}e^{\gamma L}} e^{-\alpha L}$
$I_2(0) = I_2(L) \frac{1+m}{1+me^{-\gamma L}} e^{-\alpha L}$
$m = \frac{I_1(0)}{I_2(L)}$
$\alpha \ll \gamma$
<i>Kerr media (nondegenerate codirectional coupling)</i>
$I_1(L) = I_1(0) \frac{1+m^{-1}}{1+m^{-1} \exp[(\gamma/\alpha)(1-e^{-\alpha L})]} e^{-\alpha L}$
$I_2(L) = I_2(0) \frac{1+m}{1+m \exp[-(\gamma/\alpha)(1-e^{-\alpha L})]} e^{-\alpha L}$
$m = \frac{I_1(0)}{I_2(0)}$
$\gamma = \frac{2\pi}{\lambda \cos \theta} \frac{n_2^I}{[1 + (\Omega\tau)^2]^{1/2}} [I_1(0) + I_2(0)] \sin \varphi$
$\varphi = -\tan^{-1}(\Omega\tau)$

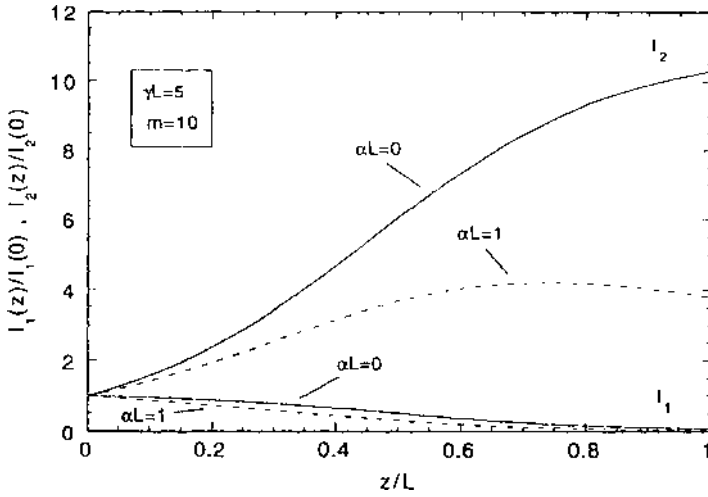


Figure 18 Intensities I_1 and I_2 as functions of z for two-beam coupling in a photorefractive medium.

$m = I_1(0)/I_2(0)$. The behavior of these solutions is shown in Fig. 18. Beam amplification requires that $\gamma > \alpha$. For large m , $I_2(z) \approx I_1(0)\exp[(\gamma - \alpha)z]$. The amplification of beam 2 is given by a gain factor G :

$$G = \frac{I_2(L)}{I_2(0)} = \frac{1 + m}{1 + m \exp(-\gamma L)} e^{-\alpha L} \quad (66)$$

In contradirectional beam coupling, the two beams are incident on the crystal from opposite directions. Both beams are assumed to make an angle θ with respect to the z -axis. The coupled equations are nearly identical to Eqs. (62) and (63), except for I_2 the signs of γ and α are reversed. These equations cannot be solved exactly except for $\alpha = 0$. However, provided that $\alpha \ll \gamma$, the resulting approximated solutions have the identical form as those for codirectional coupling [8]. It should be noted now that the two input waves are $I_1(0)$ and $I_2(L)$, and $m = I_1(0)/I_2(L)$. Defining transmittances $T_1 = I_1(0)/I_1(L)$ and $T_2 = I_2(L)/I_2(0)$, it is noted that $T_2 = T_1 \exp(\gamma L)$. Thus, for $\gamma L \gg 1$, beam 1 traveling to the right is strongly attenuated, and beam 2 traveling to the left is amplified. This nonreciprocal transmission has no analog in linear optics and has a potential application for a one-way viewing window.

When the beams are nondegenerate (i.e., $\omega_1 \neq \omega_2$), a traveling grating is produced with velocity $V = \Omega\Lambda/2\pi$, where Λ is the grating spacing and $\Omega = \omega_2 - \omega_1$. This causes an additional phase shift, with the net phase shift

given by

$$\varphi = \varphi_0 - \tan^{-1}(\Omega\tau) \quad (67)$$

where φ_0 is the phase shift for $\Omega = 0$ and τ is the response time of the photorefractive medium. For a diffusion controlled space charge field, the two-beam gain decreases as Ω increases. This is because φ is at the optimum value of $\pi/2$ for $\Omega = 0$. When an external field is applied to optimize the space charge field (and hence the index modulation), $\varphi_0 \neq \pi/2$. Thus in this case the nondegeneracy can be exploited to drive the phase shift to $\pi/2$ and enhance the gain.

In the situations described above, the input waves were assumed to have the same polarization. When this is not the case, an anisotropic two-wave mixing can occur that mixes waves of different polarization (cross-polarization coupling) [8]. However, this coupling is generally small in birefringent materials because the optical anisotropy creates different Bragg conditions for the two independent polarization states. Cross-polarization coupling can be significant however in cubic materials, such as GaAs. The faster photorefractive response in these materials makes this case of much interest. (See Ref. 8 for more details.)

Nondegenerate two-beam coupling in Kerr media. As mentioned above, the interference of two nondegenerate waves creates a traveling interference pattern. In a Kerr medium, this produces a traveling index grating through the nonlinear index of refraction. The index pattern for codirectional beam coupling can be written as

$$n = n_0 + \Delta n_0 + 2\{n'_2 e^{i\varphi} A_1^* A_2 \exp[i(\mathbf{K} \cdot \mathbf{r} - \Omega t)] + \text{c.c.}\} \quad (68)$$

where

$$|\mathbf{K}| = |\mathbf{k}_2 - \mathbf{k}_1| = \frac{2\pi}{\Lambda} \quad (69)$$

and Δn_0 is a uniform nonlinear index change. The phase φ indicates the degree to which the index grating is temporally delayed (spatially shifted) with respect to the interference pattern and is a result of the finite material response time τ . Assuming a steady-state model in which the grating is written instantaneously but decays with a time constant τ [17],

$$n'_2 e^{i\varphi} = \frac{n_2}{1 + i\Omega\tau} \quad (70)$$

and therefore

$$\varphi = -\tan^{-1}(\Omega\tau) = -\tan^{-1}\left(2\pi \frac{V\tau}{\Lambda}\right) \quad (71)$$

while $n_2' = n_2/(1 + \Omega^2\tau^2)^{1/2}$. Thus if the distance traveled by the grating in a time τ is large compared to Λ , then $\varphi \rightarrow -\pi/2$.

The solutions to the coupled wave equations are given in Table 11, where m is again the input intensity ratio. Notice the similarity to the solutions for two-beam coupling in a photorefractive medium. Hence again there is a nonreciprocal energy transfer from one wave to the other. The coupling coefficient in this case is given by

$$\gamma = \frac{2\pi}{\lambda \cos \theta} \frac{n_2'}{[1 + (\Omega\tau)^2]^{1/2}} [I_1(0) + I_2(0)] \sin \varphi \quad (72)$$

where λ is the mean wavelength. Notice in this case that the coupling coefficient depends on the total input intensity, unlike the photorefractive case, for which the coupling coefficient depends on the intensity ratio. Also note that the coupling is zero if $\varphi = 0$. Thus there is no energy transfer between the waves when the frequencies are degenerate, in which case there is no phase shift between the grating and the interference pattern.

In the lossless regime ($\alpha = 0$), the transmittances of the two beams are

$$T_1 = \frac{I_1(L)}{I_1(0)} = \frac{1 + m^{-1}}{1 + m^{-1} \exp(\gamma L)} \quad (73)$$

$$T_2 = \frac{I_2(L)}{I_2(0)} = \frac{1 + m}{1 + m \exp(-\gamma L)} \quad (74)$$

The behavior of these solutions is illustrated in Fig. 19. For $n_2 > 0$, the low-frequency wave will always see gain at the expense of the high-frequency wave.

As pointed out by Yeh [17], there is no cross-coupling of the phases in nondegenerate two-beam coupling in Kerr media. Thus a phase aberrated beam can be used to amplify a “clean” beam without introducing any phase distortions.

Two-beam coupling of chirped pulses in Kerr media. A particular case of nondegenerate two-beam coupling occurs in time-delayed two-beam experiments using chirped pulses. Pulses exhibiting linear frequency chirp have an instantaneous frequency given by $\omega(t) = \omega_0 + bt$, where ω_0 is the center frequency of the pulse and b is the linear chirp coefficient. For $b > 0$, the leading edge of such a pulse is red-shifted from the center frequency (obtained at the peak of the pulse) while the trailing edge is blue-shifted. Consequently, when one pulse is delayed in time relative to the other pulse, the instantaneous frequencies of the two pulses are nondegenerate where they overlap spatially. The coupling between two such pulses can be described by equations analogous to those given above, in terms of integrated pulse energy instead of instantaneous intensity, with the substitution $\Omega \rightarrow 2b\tau_d$, where τ_d is the delay time between the two pulses. As the delay time is swept from positive to negative values, the sign of the coupling coefficient changes sign. Thus, energy flows first from one pulse to the other, then

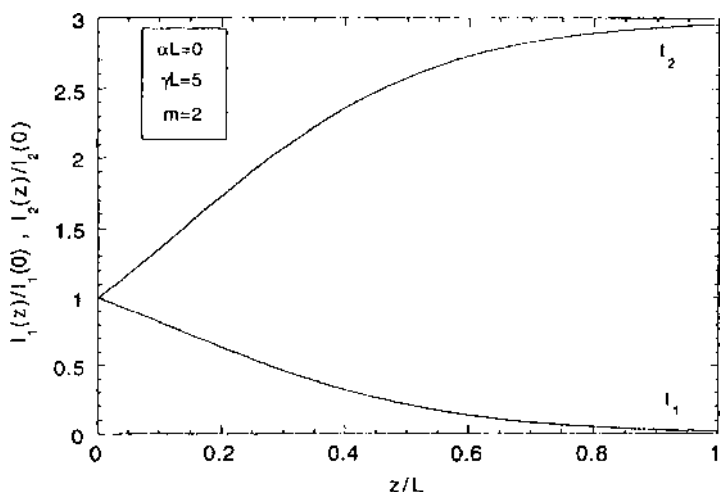


Figure 19 Intensities I_1 and I_2 as functions of z for nondegenerate two-beam coupling in a Kerr medium.

reverses as the delay between them is reversed. The energy flow is always from the high-frequency beam to the low-frequency beam as long as both $n_2 > 0$ and $b > 0$.

Applications. Several applications have been explored utilizing two-beam coupling. Some of these are listed in Table 12. Although most of these have exploited the large gain in photorefractive media, the flexible rejection filter has been investigated using Kerr media as well. Such a filter can be used for protecting optical sensors from high-intensity lasers. By using the codirectional

Table 12 Some Applications of Two-Beam Coupling

Flexible rejection filter
“One way” window
Photorefractive resonators (self-adjusting phase)
Optical computing
Optical interconnects
Optical neural networks
Image amplification
Laser beam clean-up
Spatial light modulators

beam coupling described above, the filter selectively responds to coherent radiation by transferring the laser beam energy to a beam dump while the weak beam is directed to the sensor's detector. Since two-beam coupling is a coherent process, the filter does not respond to ordinary scene radiation which is broadband and incoherent. Other applications listed in Table 12 are more thoroughly discussed in the literature [8].

D. Optical Bistability

The first optical bistability experiments were performed using a Fabry–Perot interferometer with a nonlinear material situated between the two mirrors of the cavity [18]. Much of the interest in this phenomenon has centered around this nonlinear étalon.

A Fabry–Perot étalon is a narrow-band transmission filter. The spectral location of the transmission peak can be swept in frequency, or wavelength, by varying the optical path length between the cavity mirrors. This can be accomplished by varying the physical distance between the mirrors, or by varying the index of refraction of the medium between the mirrors. If the medium has an intensity dependent index of refraction, then light can be used to tune the cavity.

Steady-state optical bistability in a Fabry–Perot. The simplest Fabry–Perot consists of two partially transmitting mirrors separated by a distance L and filled with a dielectric material. The intensity of light I_t transmitted by this device for an incident intensity of I_i is given by

$$\frac{I_t}{I_i} = \frac{T^2}{T^2 + 4R \sin^2 \varphi / 2} \quad (75)$$

where T and R are the mirror transmittance and reflectance, respectively, and $\varphi = 4\pi nL/\lambda$. If $n = n_0 + n_2^I I_c$, where I_c is the internal cavity intensity, then this phase becomes intensity dependent. The cavity intensity is related simply to the device transmitted intensity by $I_t = TI_c$. Thus

$$\varphi = 4\pi n_0 \frac{L}{\lambda} + \frac{(4\pi L/\lambda)n_2^I}{T} I_t = \varphi_0 + 2aI_t \quad (76)$$

The equation for the transmitted light intensity from a nonlinear Fabry–Perot is shown in Table 13.

This solution for I_t is a transcendental equation and cannot be simplified. A graphical solution is illustrated in Fig. 20. The curve in this figure is the Fabry–Perot Airy function [19] and depends on I_t . The straight lines are of the form $y = I_t/I_i$, i.e., they have zero intercept and are parameterized by the slope I_i^{-1} . Thus a vertical line would correspond to $I_i = 0$, and as $I_i \rightarrow \infty$ the line becomes horizontal. The solution for any given I_i is the intersection of the straight line with

Table 13 Formulas Related to Optical Bistability

Nonlinear dispersive Fabry–Perot étalon (Kerr-like media)

Optical bistability

$$\frac{I_t}{I_i} = \frac{T^2}{T^2 + 4R \sin^2\left(\frac{1}{2}\varphi_0 + aI_t\right)}$$

$$I_i = I_t \left[1 + \frac{4R}{T^2} \sin^2\left(\frac{1}{2}\varphi_0 + aI_t\right) \right]$$

$$a = \frac{2\pi L n_2'}{\lambda T}$$

Differential gain

$$\frac{dI_t}{dI_i} \simeq \left[1 + 4a \left(\frac{R}{T}\right)^2 (2aI_t + \theta) I_t + \left(\frac{R}{T}\right)^2 (2aI_t + \theta)^2 \right]^{-1}$$

$$\left(\frac{dI_t}{dI_i}\right)_{\max} \simeq \left[1 - \left(\frac{R}{T}\right)^2 \theta^2 / 3 \right]^{-1} \text{ at } I_t = -\theta / 3a$$

Nonlinear dispersive distributed feedback system (Kerr-like media)

$$J_i = y_3(J_i) + \frac{y_3(J_i) - y_2(J_i)}{[y_1(J_i) - y_2(J_i)][y_1(J_i) - y_3(J_i)]^{-1} \text{sn}^2(u, \gamma) - 1}$$

$$y_1 > y_2 > y_3 > y_4 = \text{roots of } Q(y) = 0, \text{ Eq. (80)}$$

$$u = 2[(y_1 - y_3)(y_2 - y_4)]^{1/2}$$

$$\gamma = \frac{(y_1 - y_2)(y_3 - y_4)}{(y_1 - y_3)(y_2 - y_4)}$$

$$y = I_t / I_c \quad J_i = I_i / I_c \quad J_c = I_t / I_c$$

$$I_c = \frac{2\varepsilon_0 n_0^2 c \lambda}{3\pi n_2 L} = \frac{2n_0 \lambda}{3\pi n_2^2 L}$$

Mixed absorptive and dispersive ring cavity (two-level system) (ℓ_{NL} = length of nonlinear medium in cavity)

$$I_i = I_t \left\{ \left[1 + \frac{\alpha_{00} \ell_{\text{NL}}}{2T} \frac{1}{1 + (T_2 \Delta)^2 + I_t / T I_s^\Delta} \right]^2 \left[\frac{\alpha_{00} \ell_{\text{NL}}}{2T} \frac{T_2 \Delta}{1 + (T_2 \Delta)^2 + I_t / T I_s^\Delta} - \frac{\theta R}{T} \right]^2 \right\}$$

the Airy function. Note that for some incident intensities, the output is multivalued for a single input intensity. When two of these states are stable, i.e., they settle into a nearby steady state when subjected to minor perturbations, the system is said to be bistable. The intensity regime of these states, if indeed two states rather than just one exist, depends on the original phase of the system at low intensities, i.e., on φ_0 .

For the situation depicted in Fig. 20, as the incident intensity increases, the output intensity increases only slightly (points a and b), since the filter is detuned

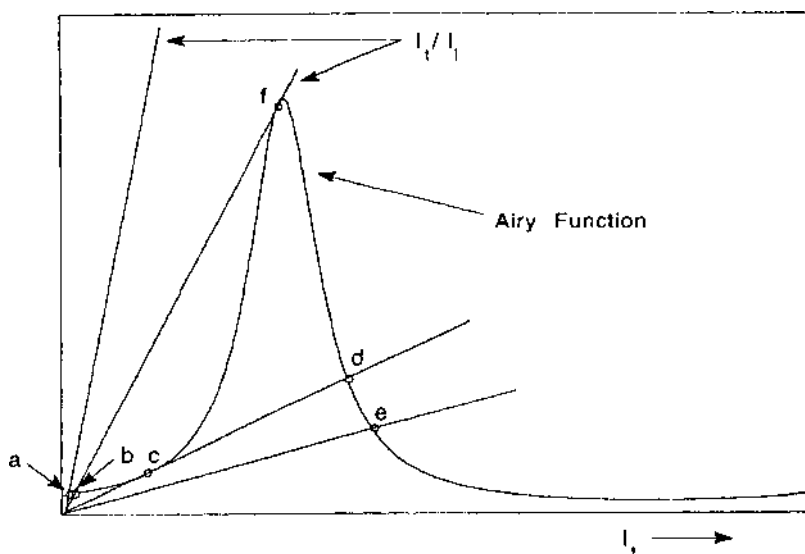


Figure 20 A graphical solution of intensity transmitted through a nonlinear Fabry–Perot étalon, illustrating optical bistability.

from resonance. At point c, a minor increase in I_i causes the output to jump to a high value. This is called optical switching. For further increases in I_i , the transmitted intensity increases only slightly. This is a power limiting region, since the increasing light intensity is tuning the filter away from resonance, decreasing its transmittance. As I_i is now decreased, the output remains high until the point f is reached. This is the region of bistability. Since the device has been put in a high transmittance state, the internal cavity intensity is high enough to keep it there over a limited range of input intensity. A minor drop of input intensity at point f causes the device to switch down to low transmittance. Thus as the incident intensity is ramped up and down, the device output traces out a hysteresis curve, which has come to be the signature of optical bistability.

It is also possible to obtain a graphical solution by plotting the equation in Table 13 as a function $I_t(I_i)$, with I_i as the independent variable, and then inverting the axes. This is illustrated in Fig. 21, where the solutions are given for various values of ϕ_0 . The three cases shown illustrate the three different operating conditions of the nonlinear Fabry–Perot: (a) power limiting, (b) differential gain, and (c) optical bistability. The negative slope region in Fig. 21c has been shown to be unstable [20]. Therefore the output makes discontinuous jumps at these turning points as discussed above.

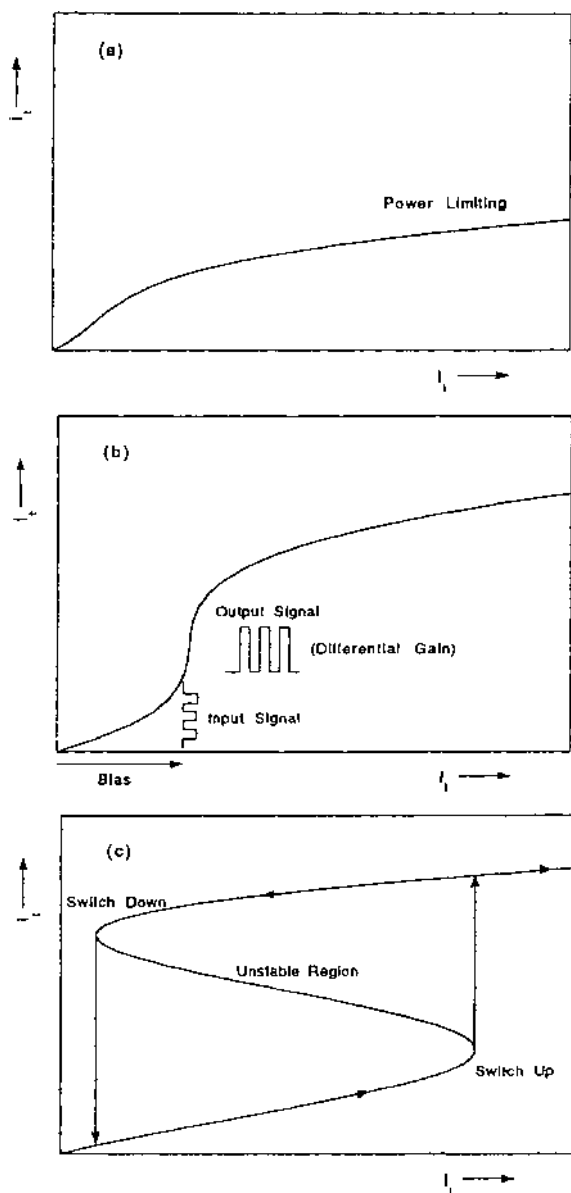


Figure 21 Transmitted intensity as a function of incident intensity for a nonlinear Fabry-Perot étalon for various detunings from resonance. (a) Power limiter; (b) optical transistor; and (c) optical switching and bistability.

The differential gain region has been proposed as an optical transistor. If the system is biased by a light beam so that the output is near the knee of this curve, then a small variation in this beam, or an input by another beam, will result in a relatively large output variation. This is illustrated in Fig. 21b.

Further insight into the differential gain and bistable regions can be obtained by an approximate analysis of the system when it is near resonance [18]. The relation between the input and output fields, E_i and E_t , for the Fabry–Perot is

$$E_i = \frac{1 - Re^{i\varphi}}{T} E_t \quad (77)$$

Take the phase to be $\varphi = 2M\pi + \theta + 2aI_t$, where M is an integer and $\theta + 2aI_t \ll 2M\pi$. Thus θ is the original detuning of the Fabry–Perot from resonance. Expanding the exponential in Eq. (77) to lowest order in θ , taking the derivative of this expression, and inverting yields the result for the differential gain dI_t/dI_i shown in Table 13. The differential gain will have the maximum value shown when the input intensity is such that $I_t = -\theta/3a$. This maximum gain will approach infinity as $(\theta R/T)^2 \rightarrow 3$. Thus the cavity detuning and finesse (i.e., mirror reflectance) set the conditions for the differential gain mode $[(\theta R/T)^2 \leq 3]$ or the optical bistability mode $[(\theta R/T)^2 > 3]$.

Optical bistability in distributed feedback systems. The minimum requirements for optical bistability are illustrated in the Fabry–Perot device: a nonlinear transmission with positive feedback. In a Fabry–Perot, the feedback is provided at two points: the cavity mirrors. In a Bragg filter, for example, a hologram, the feedback is distributed throughout the device. If the material is nonlinear, this type of system will also exhibit optical bistability as well as power limiting and differential gain [21].

In the steady-state analysis of the Fabry–Perot device, the cavity intensity was taken as a constant between the mirrors. Ignoring the spatial variation of the standing wave gives what is sometimes called the mean field approximation. Gibbs [20] discusses some of the effects of standing waves. However, the mean field approximation is adequate to analyze the main properties of the device.

For a distributed feedback (DFB) device, standing wave effects are still ignored. The envelope of the internal intensity is, however, not spatially uniform in these types of systems. This spatial nonuniformity cannot be ignored, so the analysis is somewhat more involved [21].

The index distribution for a nonlinear DFB device is taken to be

$$n(z) = n_0 + n_1 \cos 2Kz + 2n_2 |E(z)|^2 \quad (78)$$

where $n_1 \ll n_0$ is the amplitude of an index grating with a grating spacing of $\Lambda = \pi/K$. A coupling coefficient $\kappa = \pi n_1/\lambda$ and phase mismatch $\Delta k = k - K$ are defined, where κ couples forward and backward traveling waves of wave

vector $k = n_0\omega/c$ in the DFB device. Thus the total complex field amplitude is taken to consist of forward and backward propagating waves:

$$E(z) = A_f(z)e^{ikz} + A_b(z)e^{-ikz} \quad (79)$$

These forms for the field and index of refraction are substituted into the wave equation and result in two coupled first order differential equations for the forward and backward waves in the slowly varying amplitude approximation. These can be solved exactly, and the solution for the transmitted intensity $I_t = I_f(L)$ is given in Table 13 in terms of normalized intensities.

The normalization is in terms of a characteristic intensity

$$I_c = \frac{2\varepsilon_0 n_0^2 c \lambda}{3\pi n_2 L} = \frac{2n_0 \lambda}{3\pi n_2^2 L} \quad (80)$$

which sets the intensity scale for nonlinear operation. The parameters $y_1 > y_2 > y_3 > y_4$ are roots of a fourth order polynomial $Q(y)$,

$$Q(y) = (y - J_t)[(\kappa L)^2 y - (y - J_t)(\Delta \kappa L + 2y)^2] \quad (81)$$

where $y = I_f/I_c$, and are thus functions of the normalized transmitted intensity J_t . The solution $J_t(J_i)$ is given in terms of a Jacobi elliptic sine function, which was introduced in Chapter 2.

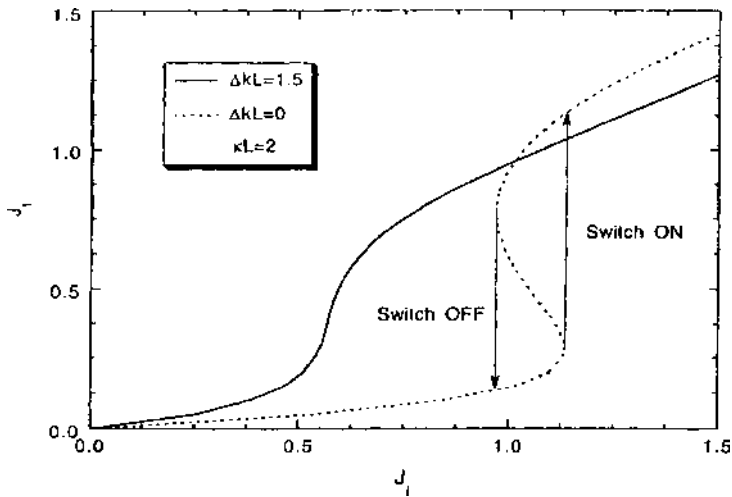


Figure 22 Transmitted intensity as a function of incident intensity of nonlinear distributed feedback device (Bragg reflector), illustrating optical bistability.

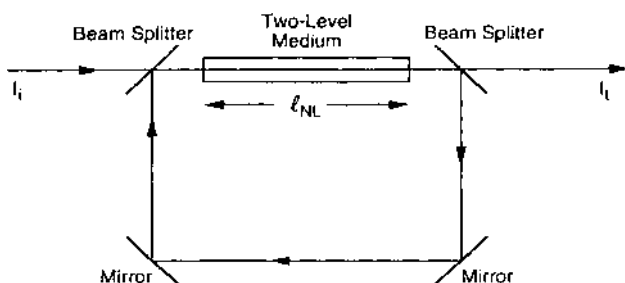


Figure 23 Ring cavity optically bistable device.

The behavior of these solutions is shown in the plot of Fig. 22. Negative slope regions are again unstable. The parameters determining regions of differential gain and optical bistability are now κL and $\Delta \kappa L$, which correspond to R and θ for the Fabry–Perot. Note that although the Bragg filter is a narrow band *reflection* filter, while the Fabry–Perot is a narrow band *transmission* filter, both can exhibit optical bistability. The key features are nonlinear transmission and positive feedback.

Mean field model of a ring cavity filled with a two-level system. Bonifacio and Lugiato [22] considered a nonlinear ring cavity as illustrated in Fig. 23, where the nonlinear medium is a two-level system (e.g., sodium vapor near resonance). Using the terms defined for two-level systems in Section II.E, the solution $I_i(I_t)$ to the analysis of this ring cavity system, which avoids standing-wave effects, is given in Table 13. This solution is for a homogeneously broadened two-level system. To include inhomogeneous broadening (e.g., Doppler broadening), the line shape should be integrated over a suitable distribution function (see Ref. 20).

The ring cavity/two-level medium forms a mixed bistable system. The first term in the solution $I_i(I_t)$ corresponds to that of a pure absorptive bistable system when $\Delta = \theta = 0$, where θ was defined above for a pure dispersive bistable system (no nonlinear absorption). The properties of saturable absorption are discussed further in Chapter 9. The second term corresponds to a pure dispersive bistable system when $T_2 \Delta \gg 1$. The first observations of optical bistability were performed with atomic sodium vapor in a Fabry–Perot with the dispersive part dominating [18]. The behavior of the solutions for this model are illustrated in Fig. 24.

Other approaches. Since the initial studies of a nonlinear Fabry–Perot with a two-level atomic vapor, several systems have been demonstrated to exhibit optical bistability. Some of these are listed in Table 14. Nonlinearities exploited include population redistribution in gases and semiconductors (both band filling and exciton saturation effects), Kerr nonlinearities in liquids, electrostrictive

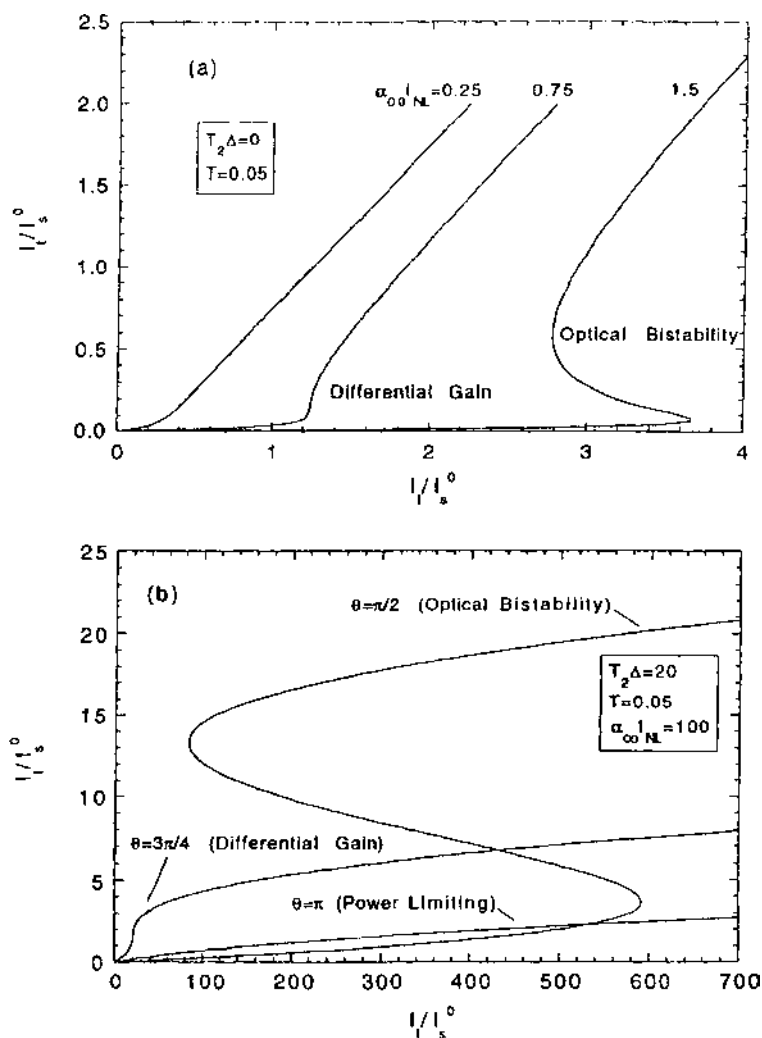


Figure 24 Transmitted vs. incident intensity for a two-level system in a ring cavity. (a) Pure absorptive system and (b) mixed absorptive and dispersive system.

effects in particle suspensions, thermal nonlinearities, electrooptic effects in solids and liquid crystals, as well as magneto-optic and acousto-optic effects. These latter devices are hybrid in the sense that the positive feedback is not obtained directly from the transmitted light but rather indirectly through an electronic signal proportional to the transmitted intensity. Several different

Table 14 Additional Optical Bistable Devices

Device	Ref.
Semiconductor étalons	
GaAs	[73–75]
GaAs–GaAlAs multiple quantum well	[76]
InSb	[77]
ZnS/ZnSe interference filters (thermal effects)	[78–80]
Color filters, dyes (Fabry–Perot)	[74,80–82]
Self-trapping, self-lensing, self-bending	[83–85]
Others not strictly related to n_2	[20]
Hybrid devices (electronic feedback)	
Pockels cell Fabry–Perot	[86–89]
Pockels cell polarization modulator	[90,91]
Pockels cell Mach–Zehnder interferometer	[92]
Pockels cell Michelson interferometer	[93]
Magneto-optic modulator	[94]
Acousto-optic modulator	[95]
Electro-optic scattering	[96]

optical phenomena have been exploited, including interference, polarization rotation, focusing, and scattering. Again, the key features are a nonlinear transmission as a function of some control variable proportional to the transmitted light intensity, and positive feedback to the system.

Transient effects. The above discussion summarizes the steady-state operation of optical bistable systems. Transient effects can arise when the system is pumped by a pulsed light source, or when pumped by a cw source and instabilities set in. A few of these effects are cited below.

Bischofberger and Shen [23] studied the effects of a nonlinear Fabry–Perot containing a Kerr medium when pumped with a pulsed laser. They examined these effects theoretically and experimentally in the power limiting, differential gain, and optical bistability regions. The effects observed depend on relations between three fundamental time constants: the round-trip cavity time for light $t_R = 2nL/c$, the response time of the nonlinearity in the medium τ_M , and the pulse width τ_p of the laser.

The basic results of the analysis of these transient effects can be summarized as follows. For $t_R \ll \tau_p \ll \tau_M$, there is a “bistability” in all regions (power limiting, differential gain, and optical bistability), but no sharp switching as the pulse ramps up and then down. This is illustrated in Fig. 25. Basically, the system response lags the optical pulse as it propagates through, and there is not enough time for a quasi-steady state to form. When $\tau_M \ll t_R \ll \tau_p$, quasi-steady

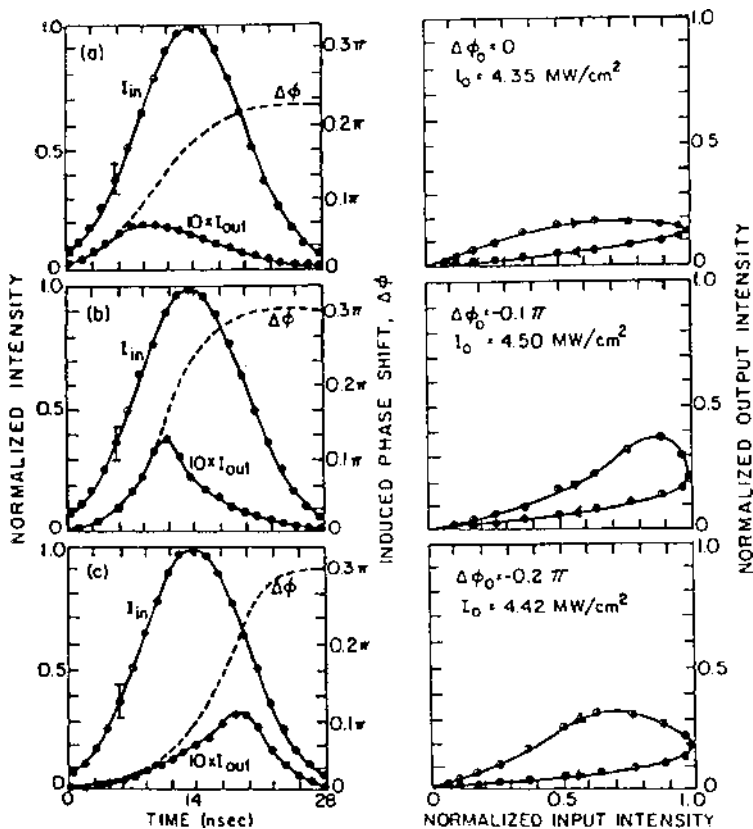


Figure 25 Transient effects in a nonlinear Fabry-Perot for $t_R \ll \tau_p \ll \tau_M$ [$t_R = 0.11$ ns, $\tau_p = 14$ ns, $\tau_M = 145$ ns (liquid crystal MBBA)]. (From Ref. 23.)

state regions of power limiting, differential gain, and optical bistability can be observed. However, in the switch up region of the bistable curve, there is overshoot and ringing. These effects are illustrated in Fig. 26. Optical switching is more effective (i.e., sharper) in cavities with high finesse, but the over-shoot and ringing are more evident [23].

Gibbs [20] reviews some of the literature covering other aspects of transient behavior associated with optical switching. This includes pulse shortening and pulse reshaping effects.

Optical bistability has some aspects that parallel those of phase transitions, such as abrupt switching and hysteresis. Another is a phenomenon called critical slowing down, first predicted by Bonifacio and Lugiato [24]. This refers to

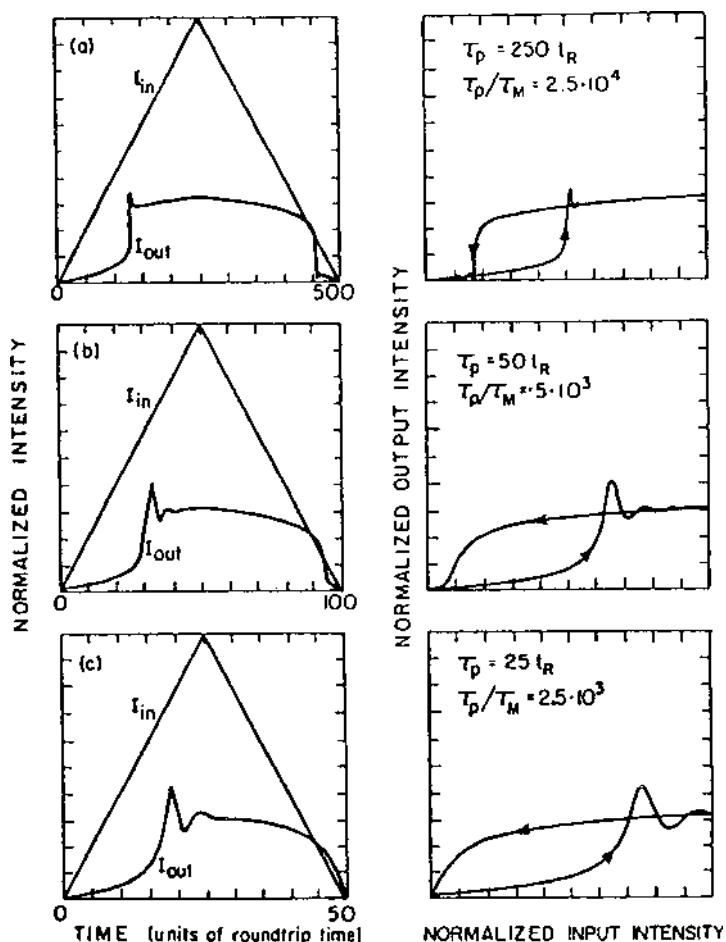


Figure 26 Computer simulations of transient effects in the switching and optical bistability behavior of a nonlinear Fabry–Perot for $\tau_M \ll t_R \ll \tau_p$ ($\tau_M = 0.01 t_R$). (From Ref. 23.)

the power or intensity required to switch a bistable device ON, and the time it takes for the device to assume this ON state. Basically, if the input intensity is much larger than the switch ON intensity corresponding to the steady state, the system responds quickly, limited only by intrinsic time restraints such as the cavity build-up time in a Fabry–Perot. However, if the input intensity is only slightly greater than the steady-state switch ON value, the system response slows down. The switch ON time actually diverges as the input intensity approaches

the steady-state switch ON value from above. This is called critical slowing down and has been observed experimentally in a hybrid system by Garmire et al. [25], in an absorptive bistable system by Grant and Kimble [26], and in a thermal dispersive system (interference filter) by Karpushko and Sinitsyn [27].

The above discussion relates transient effects to the results of pulsed inputs. Transient effects also arise due to intrinsic instabilities of the system when the input is cw. One such effect is evident when there are two nonlinear contributions to the system response of differing sign and differing time constants [28]. An example would be a slightly absorbing material that exhibits a fast, positive Kerr response and a slow, negative thermal response. In such a system, biased for optical bistability, as the cw beam is turned on, the system finds itself in the ON state after a short transient response. The high transmission state produces absorption with a slowly responding thermal effect. The opposite sign of this nonlinearity causes the operating point on the optical bistability curve to drift, and the device eventually switches OFF. The resulting low transmission allows the device to cool and reset the optical bistability operating point. When this occurs, the fast Kerr nonlinearity causes the device to switch ON again, and the entire sequence recurs. The result is regenerative pulsations in the output for a steady cw input (i.e., the system functions as a relaxation oscillator). The period of oscillations depends on the time constants involved. Gibbs [20] discusses several other cases.

Another type of instability, called the Ikeda instability [29], can set in for cw inputs with $t_R \gg \tau_M$. In this case, the nonlinear response of the medium adiabatically follows the field. The instability sets in at high intensities in the bistable region. Although Ikeda first explored this instability theoretically for dispersive optically bistable systems in cavities, the results are perfectly general to all bistable systems and were first studied experimentally in a hybrid optical bistable system [30]. This system consisted of a Pockels cell based polarization modulator with electronic feedback proportional to the output intensity, but deliberately delayed electronically to satisfy the condition $t_R \gg \tau_M$, where now t_R refers to the feedback time of the device.

The Ikeda instability can be understood as follows. The device is assumed to be in a high transmission state. As the incident intensity is increased above some threshold, it reaches a value at which it would switch to a low transmission in the steady state. However, since the feedback is slow, light does not see this state until after a delay time t_R when the transmission switches to a low value. Since the transmission is now low, the intensity is not sufficient to keep it in this state. But since the feedback is not instantaneous, the transmission is kept low for a time t_R before it switches high again. The situation is now as it started, and the process continues. The result is that the output intensity oscillates with a period $2t_R$ even though the input is cw.

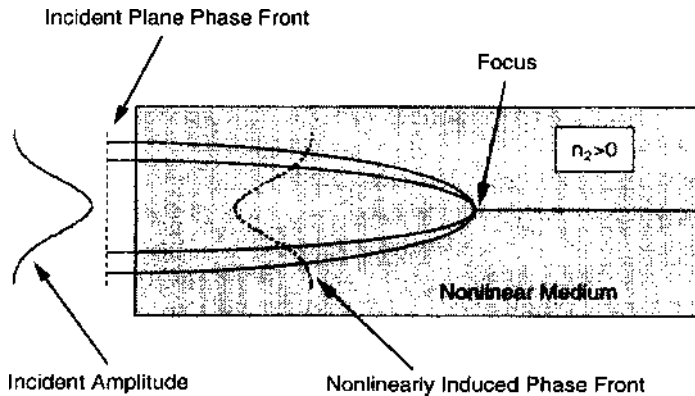


Figure 27 Self-focusing of a nonuniform beam of light in a nonlinear medium.

At a higher input intensity, another instability sets in, which produces a slightly modified oscillation pattern with a period of $4t_R$. As the system is driven even harder, the time required for the waveform to reproduce itself continues to double. So, at each bifurcation, the period doubles in the sequence $2t_R$, $4t_R$, $8t_R$, $16t_R$, ..., etc. This period doubling rapidly leads to a situation where the waveform does not repeat itself at all. This is called optical chaos. It is a phenomenon in which a completely deterministic nonlinear system exhibits apparently random behavior. Optical chaos has generated considerable interest since its behavior corresponds to phenomena in quite different systems, e.g., turbulence in fluids, fluctuations in population growth, chaotic behavior in financial markets, and apparent random behavior in biological systems.

E. Self-Focusing

Self-focusing occurs when a light beam of nonuniform intensity falls on a medium with a nonlinear index of refraction. As illustrated in Fig. 27, since the nonlinear index follows the shape of the beam (for local effects without saturation), an index gradient is induced in the medium. For a positive nonlinearity, this means that a greater index, and hence larger phase retardation, is induced on-axis than in the wings of the beam. This has the effect of creating a positive lens that tends to focus the beam.

Thin media. A wave with a plane phase front and a Gaussian amplitude distribution incident on a thin nonlinear medium will induce an index modulation

Table 15 Formulas Related to Self-Focusing in Kerr-Like Media*Thin nonlinear medium*

Nonlinear lens focal length for incident Gaussian beam with planar phase front (paraxial approximation)

$$f = \frac{n_0 w^2}{4n_2^l I_0 L}$$

Field distribution at far field point z' due to thin nonlinear medium at point z of Gaussian beam ($\Delta z = z' - z$)

$$E(z', r, t) = E(z, 0, t) \sum_{m=0}^{\infty} \frac{[i\Delta\varphi(z, t)]^m}{m!} \frac{w_{m0}}{w_m} \exp\left(-\frac{r^2}{w_m^2} + i\frac{kr^2}{2R_m} + i\theta_m\right)$$

$$\Delta\varphi(z, t) = \frac{(2\pi/\lambda)n_2^l I_0(t)}{1 + (z/z_R)^2}$$

$$w_{m0}^2 = \frac{w^2(z)}{2m + 1}$$

$$w_m^2 = w_{m0}^2 \left[g^2 + \left(\frac{\Delta z}{\Delta z_m} \right)^2 \right]$$

$$g = 1 + \frac{\Delta z}{z[1 + (z_R/z)^2]}$$

$$\Delta z_m = kw_{m0}^2/2$$

$$R_m = \Delta z \left[1 - \frac{g}{g^2 + (\Delta z/\Delta z_m)^2} \right]^{-1}$$

$$\theta_m = \tan^{-1} \left[\frac{(\Delta z/\Delta z_m)}{g} \right]$$

Thick nonlinear medium

Critical power for self-trapping ($n_2 > 0$)

$$\mathcal{P}_{\text{cr2}} = \frac{(1.22\lambda)^2 \pi \epsilon_0 c}{32n_2} \quad (\text{SI}) \quad \mathcal{P}_{\text{cr2}} = \frac{(1.22\lambda)^2 c}{128n_2} \quad (\text{cgs})$$

Paraxial approximation of critical power for catastrophic self-focusing ($n_2 > 0$)

$$\mathcal{P}_{\text{cr1}} = \frac{\epsilon_0 c \lambda^2}{8\pi n_2} \quad (\text{SI}) \quad \mathcal{P}_{\text{cr1}} = \frac{c \lambda^2}{32\pi^2 n_2} \quad (\text{cgs})$$

Self-focusing distance (beam collapse) in paraxial approximation

$$\text{Plane wave front} \quad z_{f\infty} = \frac{\pi w_0^2/\lambda}{[(\mathcal{P}/\mathcal{P}_{\text{cr1}}) - 1]^{1/2}}$$

$$\text{Curved wave front } (R = \text{radius of curvature}) \quad z_f = \frac{R}{(R/z_{f\infty}) - 1}$$

(continued)

Table 15 Continued

Whole beam self-focusing critical powers
“Weak” focus
$\mathcal{P}_1 \approx \mathcal{P}_{\text{cr1}}$
Collapsed beam
$\mathcal{P}_2 \approx \mathcal{P}_{\text{cr2}}$
Whole beam self-focusing distance (beam collapse)
$z_{\text{f}\infty} = \frac{0.369\pi w_0^2/\lambda}{(\mathcal{P}/\mathcal{P}_2)^{1/2} - 0.858}$

given by

$$n = n_0 + n_2' I_0 \exp\left(\frac{-2r^2}{w^2}\right) \quad (82)$$

where I_0 is the on-axis intensity. For distances not too far from the axis (paraxial approximation), the exponential can be expanded to lowest order:

$$n \simeq n_0 \left(1 - \frac{2n_2' I_0 r^2}{n_0 w^2}\right) \quad (83)$$

It can be shown [31] that a medium with a parabolic index profile like that in Eq. (83) will cause an incident plane wave to come to a focus. In the thin lens approximation appropriate to the assumption of a thin medium, the nonlinear focal length of this medium is given by the expression shown in Table 15. Note that the focal length decreases in magnitude with increasing on-axis intensity. Also, the lens is converging if $n_2' > 0$, and diverging if $n_2' < 0$, as expected.

If the incident beam has a finite phase curvature, then the above formulas will be less accurate. In general, a phase shift is impressed on the beam given by (for a TEM₀₀ Gaussian beam)

$$\Delta\varphi(z, r, t) = \frac{(2\pi/\lambda)n_2' I_0(t)L}{1 + (z/z_R)^2} \exp\left[\frac{-2r^2}{w^2(z)}\right] \equiv \Delta\varphi(z, t) \exp\left[\frac{-2r^2}{w^2(z)}\right] \quad (84)$$

where $w(z)$ is the z dependent beam radius, z_R is the Rayleigh range, $I_0(t)$ is the time dependent on-axis intensity, and $z = 0$ is taken at the beam waist. The field that is then propagated to a new position z' , with $\Delta z = z' - z$, is given by the set of equations in Table 15. If the phase distortion is weak, then only a few terms in the infinite sum need to be retained to calculate the resulting field amplitude and phase distribution. This set of equations is the starting point for Z-scan analysis [32], which is a measurement technique for characterizing n_2' (see Chapter 7).

Thick media ($n_2 > 0$). In thick media, the beam can actually come to focus within the medium. This can have a significant impact on the material as well as on the beam.

A simple analysis can be performed on a cw beam with a top-hat profile. The beam induces a waveguide (i.e., focusing exactly balances diffraction) at a critical power. An expression for this power, called $\mathcal{P}_{\text{cr}2}$, is given in Table 15. This is often referred to as self-trapping. Self-trapping is power dependent rather than intensity dependent since, for a given beam power, both diffraction and self-focusing are beam area dependent and tend to offset one another for $n_2' > 0$.

When the beam has a Gaussian profile, the analysis is not so simple. However, for distances not too far from the axis, a paraxial approximation can again be employed. A critical power for self-trapping (focusing balancing diffraction) can again be found, given by the expression for $\mathcal{P}_{\text{cr}1}$ shown in Table 15. Note the similarity between the two critical powers, but $\mathcal{P}_{\text{cr}1} < \mathcal{P}_{\text{cr}2}$.

For $\mathcal{P} > \mathcal{P}_{\text{cr}2}$, the beam will catastrophically focus (shrink to zero cross section) within the medium if the medium is sufficiently thick. Within the paraxial approximation, this focusing distance can be calculated, and for an incident plane wave it is shown by the expression for $z_{\text{f}\infty}$ in Table 15. (The subscript ∞ implies that the wave incident on the medium has an infinite radius of curvature.) Note that as $\mathcal{P} \rightarrow \mathcal{P}_{\text{cr}1}$, the focal distance approaches infinity in agreement with the condition of self-trapping.

If the phase of the incident wave has a finite radius of curvature R , then the corresponding focal distance is given by the expression for z_{f} given in Table 15 within the paraxial approximation. The convention is taken that $R < 0$ implies that the incident wave is converging, while $R > 0$ implies that it is diverging. Thus a converging (diverging) wave will collapse to a focus at a distance $z < z_{\text{f}\infty}$ ($z > z_{\text{f}\infty}$).

Dawes and Marburger [33] have conducted a numerical analysis of whole-beam self-focusing for Gaussian beams. They find that the phenomenon is characterized by two critical powers. The first, which they call $\mathcal{P}_1$, is just $\mathcal{P}_{\text{cr}1}$ determined in the paraxial approximation. The second, called $\mathcal{P}_2$, differs numerically by only about 1% from $\mathcal{P}_{\text{cr}2}$ determined from the top-hat beam self-trapping analysis. For incident optical power in the range $\mathcal{P}_1 < \mathcal{P} < \mathcal{P}_2$, the on-axis intensity rises as the beam comes to a “weak” focus and then falls off as the beam diffracts. When $\mathcal{P} > \mathcal{P}_2$, the beam catastrophically collapses. The focal distance for an incident plane wave is given approximately by the expression for $z_{\text{f}\infty}$ in Table 15, which applies to whole-beam self-focusing.

It should be noted that the beam collapsing to zero dimension is a mathematical concept. Physically, this does not happen. Instead, as the beam collapses, the intensity becomes so large that other nonlinear processes are triggered. This includes such phenomena as stimulated Raman scattering, dielectric breakdown, plasma formation, and permanent laser damage (if the medium is a solid). These processes rob energy from the beam and may severely distort its phase as well. This prevents the beam from further collapsing.

Self-focusing can also lead to a phenomenon known as filamentation, in which the beam becomes unstable and breaks into several tiny filaments. This occurs for beams that do not have a smooth spatial variation. Actually, this is true for nearly all beams, since a microscopic distortion of dimension Λ can become unstable if the beam power is high enough. Indeed, if the power contained within a cylinder of this dimension (residing within the beam) exceeds $\mathcal{P}_2$, that part of the beam will begin to collapse and break into a filament. This sets a critical size on the original distortion given by $\Lambda_c = (4\mathcal{P}_2/\pi I_0)^{1/2}$ for filamentation, where I_0 is the mean peak intensity of the whole beam.

When self-focusing occurs in a two-level system, a cw beam can actually collapse to a finite diameter and then propagate as a self-trapped beam. This is because as the beam intensity increases, the nonlinear index eventually saturates and cannot increase any further even if the intensity increases. Hence this acts as an internal damping mechanism, and the beam settles into a steady state with a constant diameter.

Beam collapse followed by self-trapping also *appears* to happen with nanosecond pulse propagation in a nonsaturable Kerr medium. As pointed out by Shen [3], however, what appears to be self-trapping is actually the locus of moving foci in the medium due to quasi-steady-state self-focusing. As illustrated in Fig. 28, some point on the incident pulse comes to focus first. Earlier points on the pulse have longer self-focusing distances and hence come to focus farther away and at later times. Points closer to the peak of the pulse have a shorter self-focusing distance, but arrive later than the original point. Thus the original focus appears to split into two, one part moving forward in the beam, and the other

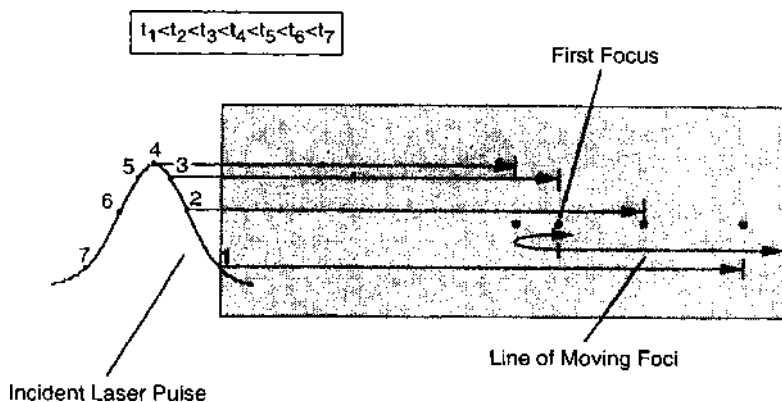


Figure 28 Schematic illustration of the track of moving foci in quasi-cw self-focusing of pulsed light.

moving backward. Eventually, the backward moving focus turns around and moves forward as points later in the pulse with longer self-focusing distances come to focus. To the eye, the moving foci appear as a continuous line, or as a self-trapped beam. As Shen has noted [3], at the turning point where the backward moving focus changes direction, the focus spends the greatest amount of time. This is the point where laser damage is likely to occur.

Thick media ($n_2 < 0$). For thick media with negative nonlinearities, the beam will not collapse but rather will diverge due to the negative lens effect. For most cases, when the temperature increases the index will decrease. Thus in a medium that absorbs part of a Gaussian beam, the index on axis is smaller than that in the wings, and the beam thus diverges. This is called thermal blooming.

Negative nonlinearities can also occur in resonant systems. For example, in a two-level system when the frequency is tuned below resonance, the index of refraction decreases due to saturation. Hence in a two-level gas, beam collapse can only be seen when the frequency is tuned to a frequency greater than the resonance frequency. In semiconductors the index coefficient is often negative, for absorption near the band edge, or for exciton saturation, or in some cases for two-photon absorption. These cases will lead to a defocusing similar to thermal blooming.

Applications. Often, self-focusing is undesirable, since it may lead to laser damage. There have been some device applications, however. The most common is optical limiting. This is illustrated in Fig. 29 for both positive and negative nonlinearities. The nonlinear medium is situated between two lenses. Low-intensity light is collected by the second lens and imaged through an aperture to a detector. High-intensity light will produce self-focusing. For $n_2 > 0$, this will result in beam collapse and phase distortion such that the light is not focused by the second lens but defocused in the aperture plane. Thus the power transmitted by the aperture to the detector is limited, and the detector is thus protected from the high-intensity radiation. Such an optical limiter has been constructed using materials like CS_2 [34]. In the negative n_2 case, the beam diverges more rapidly from the intermediate focus rather than collapsing. The result at the aperture plane is, however, the same: a defocused beam. The advantage of this type of limiter over the previous example is that it is self-protecting (i.e., the light is defocused inside the medium leading to lower intensity there that is less likely to cause laser damage). This is important when the medium is solid and the damage would be permanent. Such a limiter has been demonstrated for picosecond pulses using ZnSe as the nonlinear medium [35]. For limiting against cw and nanosecond pulses, thermal blooming in solutions with a small amount of absorbing dye has been demonstrated [36]. For such a limiter, a solvent with a high thermo-optic coefficient and low heat capacity is the best choice.

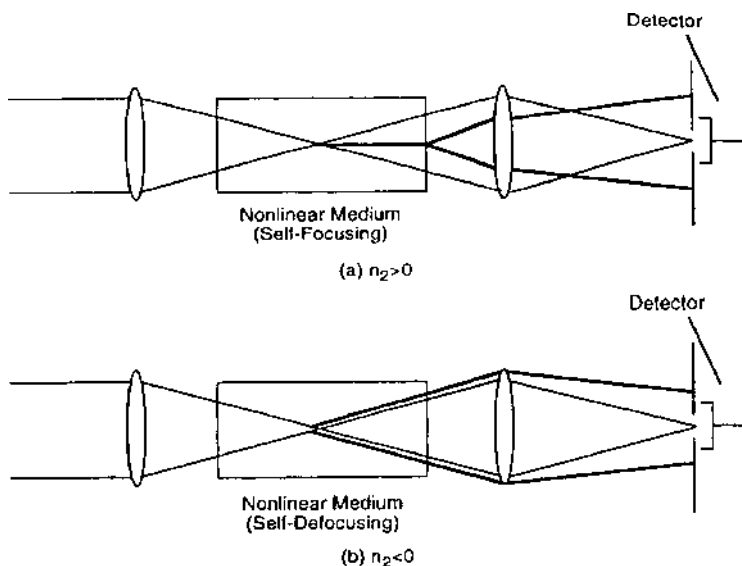


Figure 29 Optical limiters based on self-focusing. (a) $n_2 > 0$ and (b) $n_2 < 0$.

F. Beam Bending and Fanning

Other self-action effects observed with optical beams include self-bending (Kerr media) and beam fanning (photorefractive media). The self-bending in Kerr media is due to half-beam effects. In photorefractive media, whole-beam effects contribute to fanning.

Self-bending in Kerr media. In these effects, a beam block obscures half of the beam incident on a Kerr medium. For a slab beam (of infinite extent in the y -direction) with a right triangular profile in the x -direction [$I(x) = I_0(1 - x/w_0)$] that propagates in the z -direction through a thin medium, a prism is induced in the medium, which deflects the beam. The angle of deflection θ_{NL} measured in the far field is given by

$$\frac{\theta_{NL}}{\theta_d} = -\frac{\pi n_2^I I_0 L}{\lambda} \quad (85)$$

where $\theta_d = \lambda/\pi w_0$ is the far field diffraction angle of the beam. The direction of the deflected beam is to the opposite side of the axis on which the semitriangular beam is incident for $n_2^I > 0$ (positive prism). It turns out that the self-deflection angle for a slab semi-Gaussian beam is approximately equal to that given by this

equation. For a three-dimensional semi-Gaussian beam, this equation is still a fairly good approximation [37].

If the Kerr medium is thin and transparent, and the incident beam is a three-dimensional semi-Gaussian beam, then the on-axis far-field intensity is described by the relation

$$I_{\text{on-axis}} \propto \frac{\lambda w_0^4}{2\pi n_2^l L} [C^2(\Theta) + S^2(\Theta)] \quad (86)$$

where $\Theta = (4n_2^l I_0 L / \lambda)^{1/2}$. $C(\Theta)$ and $S(\Theta)$ are the Fresnel integral functions given by

$$C(\Theta) = \frac{1}{\sqrt{2\pi}} \int_0^{\Theta^2} \frac{\cos u}{\sqrt{u}} du \quad (87)$$

$$S(\Theta) = \frac{1}{\sqrt{2\pi}} \int_0^{\Theta^2} \frac{\sin u}{\sqrt{u}} du$$

Beam fanning in photorefractive media. When a single beam is incident on a photorefractive medium at some angle with respect to the c -axis of the crystal, a set of beams fans out asymmetrically from the main beam in the direction of the c -axis. This phenomenon is a whole-beam effect, not a semibeam effect as described above, and is illustrated in Fig. 30.

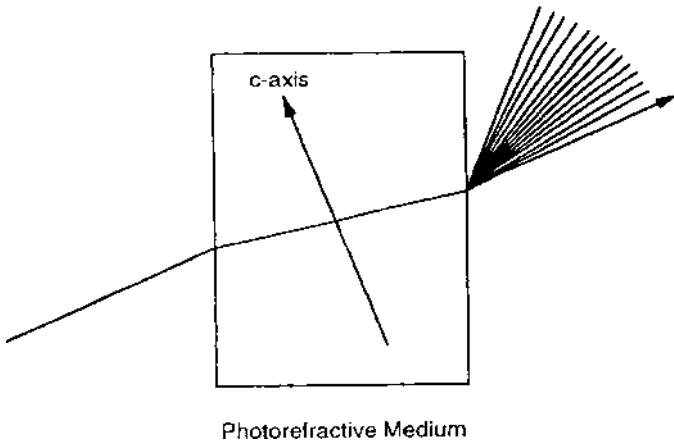


Figure 30 Schematic illustration of beam-fanning in a photorefractive medium.

Two theories have been put forth to explain this phenomenon. One, due to Feinberg [38], is a single-beam, deterministic, defocusing process. The other is a random scattering, multiple two-wave mixing process.

In the first theory, a Gaussian beam illuminates the crystal. This creates free carriers, as described above in Section II.H, which are transported by diffusion and subsequently trapped in the wings of the beam. This symmetric charge distribution leads to an asymmetric space charge field:

$$\mathbf{E}_{sc}(x, y) = \frac{-4k_B T(x\hat{x} + y\hat{y})}{e\pi w_0^2} \exp\left[-2\left(\frac{x^2 + y^2}{w_0^2}\right)\right] \quad (88)$$

$\mathbf{E}_{sc}$ produces an asymmetric refractive index profile via the Pockels effect, which creates a prism in the material. This causes the beam to be deflected asymmetrically.

Because of the tensor nature of the electrooptic effect, the beam fanning can be stronger in one dimension than in another (i.e., the fanned beam can be in the form of a sheet). The strength of the fanning can also be a function of the incident polarization.

The above analysis is valid for a thin sample and has been extended to thick media by Banerjee and Liu [39]. This model gives reasonable agreement with some experimental data and is probably accurate as one source of beam fanning in photorefractive media.

Fanning has also been hypothesized as a two-beam coupling phenomenon accompanied by weak wave amplification [40,41], where the weak wave arises due to random scattering of the incident beam from surface or volume imperfections, charge density fluctuations, etc. Some aspects of this model have been confirmed experimentally by Zhang et al. [42].

The scattered wave travels out in a different direction from the incident beam. Since the two waves are coherent, they interfere and write a photorefractive grating in the usual manner. The two waves are coupled due to the phase shift of the grating with respect to the interference pattern that recorded it. Waves scattered in the direction of the c -axis are amplified. As the scattered beam grows from noise, the modulation depth of the grating increases, which causes the two-beam coupling gain to increase. After many scattering events, a broad fan of beams develops and robs energy from the main beam.

Segev et al. [43] have extended the photorefractive two-beam coupling analysis presented earlier to the pair-wise coupling of N beams. The coupling coefficients depend on the angle between the beams and the c -axis as well as r_{eff} for the propagation direction, crystal orientation, and polarization. Noise is added in their model phenomenologically. The system of equations (sometimes for > 20 waves) is solved numerically, and the results show good agreement with several

beam fanning experiments [43]. This model has also been extended to describe the temporal evolution of beam fanning [44].

Applications. Beam fanning is sometimes detrimental, and some techniques have been developed to suppress it [45]. However, it can be used to advantage in some applications. A self-fanned beam that undergoes total internal reflection at the corner of the photorefractive crystal will propagate in the proper direction to achieve four-wave mixing with only one input beam. This leads to the concept of a self-pumped phase conjugate mirror [46]. Another application is the coherent beam excisor. Only coherent light will fan out, so this process discriminates coherent light from an incoherent background. The fanning is asymmetric and for large two-beam coupling gains can direct most of the energy in a coherent beam in one direction to a beam dump. This excises the beam and creates an optical power limiter for protection of sensitive optical sensors from harmfully high laser intensity [46].

Optical limiting by beam self-bending in Kerr media has also been discussed [47]. In this case, however, half of the incident beam must be masked to produce the asymmetry necessary for self-bending.

G. Nonlinear Phase Modulation and Solitons

Self-phase modulation (SPM). When an intense optical pulse propagates through a nonlinear medium, it gains an additional phase due to the nonlinear index of refraction. This was seen above in the optical Kerr effect, four-wave mixing, etc. If the intensity is time dependent (i.e., a short pulse), then the self-induced phase is also time dependent. For an ultrashort pulse with a center frequency ω_0 , the nonlinear phase shift is written as

$$\varphi_{NL}(t) = -\frac{\omega_0}{c} n_2^I I(t) L \quad (89)$$

The negative sign is inserted for convenience. If the material exhibits absorption at the wavelength of interest, then let $L \rightarrow L_{\text{eff}} = (1 - e^{-\alpha L})/\alpha$.

If the pulse envelope is slowly varying, an instantaneous frequency shift can be introduced by $\omega(t)t = [\omega_0 + \delta\omega(t)]t$, where $\delta\omega(t) = d\varphi_{NL}/dt$. The leading edge of the pulse has a negative slope, the peak has a zero slope, and the trailing edge of the pulse has a positive slope. Thus, the frequency shift near the peak of the pulse is zero. The leading edge is red-shifted while the trailing edge is blue-shifted. This is illustrated in Fig. 31.

Note that since $\varphi_{NL}(t)$ follows the pulse, φ_{max} occurs at the peak of the pulse. We can write $\varphi_{\text{max}} = \gamma \mathcal{P}_0 L_{\text{eff}} = L_{\text{eff}}/L_{\text{NL}}$, where $\gamma = \omega_0 n_2^I / c A_{\text{eff}}$. $\mathcal{P}_0$ is the peak power of the pulse, and A_{eff} is an effective area; for a Gaussian beam it would be $\pi w^2/2$, and for a guided wave it would be the effective mode area given

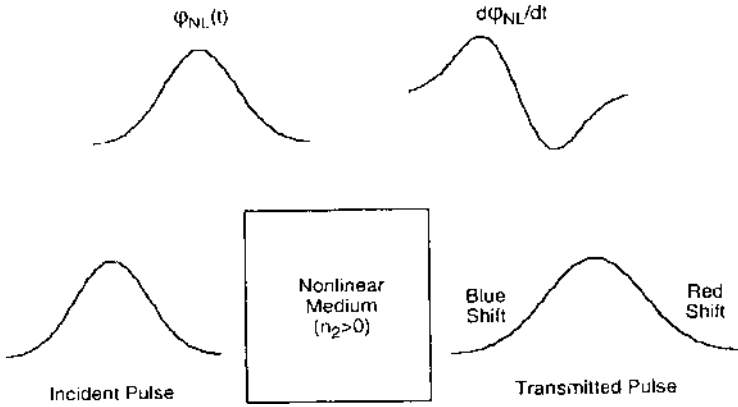


Figure 31 Self-phase modulation and frequency shifts in a nonlinear medium irradiated with a short optical pulse.

by

$$A_{eff} = \frac{\left[\int \int |a(x, y)|^2 dx dy \right]^2}{\int \int |a(x, y)|^4 dx dy}$$

where $a(x, y)$ is the normalized mode distribution across the waveguide. $L_{NL} = (\gamma \mathcal{P}_0)^{-1}$ is the effective propagation distance for which $\phi_{max} = 1$ radian.

If the pulse has a rise and fall time of τ_0 , then the maximum frequency shift is $\delta\omega_{max} \approx \Delta\phi_{max}/\tau_0$ and $\Delta\phi_{max}$ corresponds to the difference between the peak and valley in the frequency plot of Fig. 31. Self-phase modulation becomes important when $\Delta\phi_{max} \geq 2\pi$. For example, for radiation of mean wavelength $1 \mu\text{m}$ propagating through a medium of length 1 cm with $n_2^I \sim 10^{-14} \text{ cm}^2/\text{W}$, this would require a peak intensity of $> 10 \text{ GW/cm}^2$. This is not unreasonable for picosecond and sub-picosecond pulses.

The instantaneous frequency shift $\delta\omega(t)$ implies that there is a frequency chirp impressed upon the pulse that increases with L_{eff} . A self-modulated pulse will have a broadened spectrum of frequencies. The spectrum is given by the Fourier transform of the phase-modulated pulse and the extent of spectral broadening depends on the shape of the pulse. A Gaussian pulse, for example, with intensity time dependence $\exp[-(t/\tau_0)^2]$, would experience a frequency chirp given by

$$\delta\omega(t) = \frac{2t}{\tau_0^2} \frac{L_{eff}}{L_{NL}} \exp[-(t/\tau_0)^2]. \quad (90)$$

Note that the chirp has the same sign as n_2^I and is approximately linear over the central region of the pulse. For nearly square (super Gaussian) pulses, the chirp is considerably larger near the leading and trailing edges of the pulse and approximately zero over the central pulse region.

As $\mathcal{P}_0$ increases, the broadened pulse eventually displays an oscillatory structure in the frequency domain, with a number of peaks that is nearly proportional to $\varphi_{\max}$. Qualitatively, this is due to constructive and destructive interference of different points in the pulse that have the same chirp but different phase. Note that SPM can enhance or decrease chirp that is already present in the pulse.

If the medium through which the pulse passes has normal group velocity dispersion (GVD), then the pulse will also spread out in time. This is true regardless of SPM, but the nonlinear interaction increases the effect. To analyze these effects, a unitless parameter is introduced:

$$N^2 = \frac{\gamma \mathcal{P}_0 \tau_0^2}{|k_2|} = \frac{L_D}{L_{NL}} \quad (91)$$

where

$$k_2 = \left(\frac{d^2 k}{d\omega^2} \right)_{\omega_0} \quad (92)$$

is the group velocity dispersion coefficient, and $L_D = \tau_0^2/|k_2|$ is the dispersion length over which pulse spreading by GVD becomes significant. SPM can enhance pulse broadening due to GVD ($k_2 > 0$, normal dispersion) or inhibit it ($k_2 < 0$, anomalous dispersion). In the latter case, a Gaussian-shaped pulse initially broadens and then evolves into a sech^2 pulse, characteristic of the fundamental soliton (see below).

For $N \ll 1$ SPM dominates, but can enhance GVD effects because of the frequency chirp impressed on the pulse. In regions of normal GVD, a nearly rectangular pulse can be formed with an approximately linear chirp across the pulse. The pulse develops fine structure (oscillations) near its edges because of GVD. This is called optical wave breaking. For $k_2 > 0$, red-shifted light travels faster and catches up with blue light. The two different frequency components are coherent and interfere, creating oscillations. This process can be understood as four-wave mixing: fields at frequencies ω_1 and ω_2 interact via the third order polarization and create waves at new frequencies $2\omega_1 - \omega_2$ and $2\omega_2 - \omega_1$ in the pulse wings. Side lobes show up in the pulse frequency spectrum.

Additional phenomena appear for ultrashort pulses in the highly nonlinear regime. One of these is an effect called self-steepening. This involves higher order dispersion and depends on the first derivative $\partial P_{NL}/\partial t$ of the nonlinear polarization, which is usually ignored for most other process discussed in this

book. An intensity dependent group velocity is introduced, which causes the peak of the pulse to travel slower than the pulse wings. This pushes the peak toward the trailing edge of the pulse.

SPM effects can be deleterious in some circumstances. For example, in high-speed optical data transmission pulse broadening can produce a smearing out of individual bits, leading to higher bit error rates. However, in certain cases SPM can be put to advantage.

One application of self-phase modulation is super continuum generation [48]. This is useful for transient time-resolved spectroscopy. In such an experiment, an intense ultrashort pulse is split into two parts. One is incident on the sample under study and excites it by absorption. The other pulse is focused into a transparent nonlinear medium, such as a heavy water mixture or ethylene glycol. The output is frequency broadened over a wide spectral range (as much as > 100 nm), but remains as a relatively short pulse. This output pulse is time-delayed relative to the pump pulse that excited the sample, and probes the resulting transmission of the sample. The transmitted pulse is then dispersed by a grating and recorded, for example with a CCD camera. The result is a complete excited state absorption spectrum obtained in a single laser shot. The dynamics of the excited state can then be monitored by repeating the experiment as a function of delay time between the pump and super continuum probe pulses.

Cross-phase modulation. When two or more waves interact in a third order medium, the phase of one wave can be modulated by the intensity of the other wave(s). This is called cross-phase modulation (XPM). The interacting waves can be of different frequencies, or of the same frequency but orthogonal polarization. The latter effect was treated previously under the Kerr effect or polarization ellipse rotation. Here, the effects of nondegeneracy and propagation in a medium that has a linear birefringence are discussed.

When the dominant contribution to the third order polarization in a centrosymmetric medium is electronic in origin, the three independent components of the third order susceptibility are nearly equal. Under these conditions, XPM induces an index modulation of the two orthogonal components (E_x, E_y) of an input wave, given by

$$\Delta n_x = 2n_2(|E_x|^2 + \frac{2}{3}|E_y|^2) \quad (93)$$

$$\Delta n_y = 2n_2(|E_y|^2 + \frac{2}{3}|E_x|^2)$$

Consider propagation in a uniaxial medium. If the polarization of the input wave is close to the direction of the fast axis (smaller index), then the nonlinear contribution to the birefringence decreases the net birefringence relative to

the intrinsic value. As the input power is increased, it is possible for the nonlinear birefringence to cancel the intrinsic birefringence or even change the sign of the net birefringence. Thus large changes to the output polarization state (and large changes in intensity transmitted through a polarizer) are possible for small changes in input power or input polarization state. This is called polarization instability. It occurs when the input peak power is large enough to make $L_{NL} \sim L_B$, where $L_B = 2\pi/\Delta k = 2\pi/(k_{\text{slow}} - k_{\text{fast}})$ is called the intrinsic polarization beat length. A critical power $\mathcal{P}_c$ is defined at which the effective beat length goes to infinity, and is given by

$$\mathcal{P}_c = \frac{3}{2} \frac{\Delta k}{\gamma} \quad (94)$$

For small fluctuations about $\mathcal{P}_c$ the effective beat length can change dramatically. This instability can lead to chaotic output polarization if the intrinsic birefringence is periodically modulated. Such modulation can occur, for example, in an optical fiber during fabrication by rocking the preform or periodically inducing stress.

Now consider the interaction of two waves of the same polarization but at different frequencies. For cw beams or sufficiently long pulses, the steady-state form of the complex wave amplitudes exhibits a constant magnitude (meaning no energy transfer between the waves), but a nonlinear phase, which is given by

$$\begin{aligned} \varphi_{NL1} &= \frac{\omega_1 n_2^I}{c} (I_1 + 2I_2)L \\ \varphi_{NL2} &= \frac{\omega_2 n_2^I}{c} (I_2 + 2I_1)L \end{aligned} \quad (95)$$

The accumulated phase in each beam depends on the intensity in both beams. It turns out, however, that this steady state can be unstable with respect to weak, time dependent perturbations. Under certain conditions, weak perturbations can experience exponential gain as the wave propagates leading to substantial changes in the magnitude of the wave amplitude. This effect is called modulation instability.

Analysis of these perturbations requires the time dependent wave equation for an ultrashort pulse, including the effects of GVD. The necessary condition for modulation instability can be written as

$$(\Omega_1^2/\Omega_{c1}^2 \pm 1)(\Omega_2^2/\Omega_{c2}^2 \pm 1) < 4 \quad (96)$$

where Ω_j ($j = 1, 2$) are the frequencies of the perturbations (typically in the THz regime) on each wave, the $\pm$ sign corresponds to whether

the corresponding GVD coefficient is positive or negative, and

$$\Omega_{cj} = \sqrt{\frac{4\gamma_j \mathcal{P}_j}{|k_{2j}|}} \quad (97)$$

Over the range of frequencies satisfying the relation in Eq. (96), perturbations will experience a power dependent gain. If the perturbations are purposefully launched as weak probe beams at frequencies Ω_1 and Ω_2 , then the cw or long pulses will exhibit temporal modulations with period $2\pi/\Omega_j$. This can be considered a nondegenerate four-wave mixing process with gain, as discussed in Section III.B, with XPM providing the phase matching. The perturbations can also originate from spontaneous emission or thermally generated photons. In this case, perturbations at frequencies that correspond to maxima in the gain spectrum will build up most rapidly.

Finally, notice that the effects of both SPM and XPM will be present simultaneously when more than one wave is present. For ultrashort pulses this can lead to increased spectral broadening over that due to SPM alone, as well as asymmetric spectral broadening and asymmetric temporal changes to a symmetric input pulse.

Solitons. A steady-state optical wave that undergoes self-phase modulation while propagating in a medium with anomalous group velocity dispersion is unstable when subject to small perturbations [49]. The wave breaks into a string of periodic pulses. This is modulation instability as described above, except in the case of SPM it only occurs in the region of anomalous GVD. The growth of such instability by SPM is not possible under normal GVD, and thus waves in this regime are stable against weak perturbations.

In the negative group velocity dispersion region (e.g., for wavelengths $> 1.3 \mu\text{m}$ in fused silica optical fibers), there exists a condition where the propagation of a single short pulse is very stable. In fact, the pulse will propagate with constant shape and amplitude over essentially unlimited distance, limited only by linear adsorption in the medium. Such a pulse is called an optical soliton.

The nonlinear wave equation for an optical pulse propagating in the z -direction is shown in Table 16. The second derivative with respect to time of the slowly varying amplitude is included for group velocity dispersion effects. The time variable is written in terms of the retarded time $\tau = t - z/v_g$, where v_g is the group velocity of the pulse. This equation is often called the nonlinear Schrödinger equation.

A solution admitted by this equation is the fundamental soliton solution given in Table 16, where $t_{\text{FWHM}} = 1.76\tau_0$ is the width of the input pulse. This hyperbolic secant pulse will propagate without spreading or diminishing in amplitude provided the amplitude and phase are given by the expressions in

Table 16 Formulas Related to Optical Solitons

Optical solitons

Nonlinear Schrödinger equation

$$\frac{\partial A}{\partial z} + \frac{i}{2} k_2 \frac{\partial^2 A}{\partial \tau^2} = i \frac{2\omega_0 n_2}{c} |A|^2 A$$

$$k_2 = \left(\frac{d^2 k}{d\omega^2} \right)_{\omega_0} = \left(-\frac{1}{v_g^2} \frac{dv_g}{d\omega} \right)_{\omega_0}$$

$$\tau = t - z/v_g$$

Fundamental soliton solution

$$A(z, \tau) = A_0 \text{sech}(\tau/\tau_0) \exp(i\kappa z)$$

$$|A_0|^2 = -\frac{k_2}{\gamma \tau_0^2} = -\frac{k_2 c}{2n_2 \omega_0 \tau_0^2}$$

$$\kappa = -\frac{k_2}{2\tau_0^2}$$

Spatial solitons

Nonlinear Schrödinger equation

$$\frac{\partial A}{\partial z} - \frac{i}{2k} \frac{\partial^2 A}{\partial x^2} = i \frac{2n_2 k}{n_0} |A|^2 A$$

Fundamental spatial soliton solution

$$A(x, z) = A_0 \text{sech}(x/w) \exp(i\kappa z)$$

$$|A_0|^2 = \frac{n_0}{2n_2} \left(\frac{1}{kw} \right)^2$$

$$\kappa = \frac{1}{kw^2}$$

the table. Note that this means that k_2 and n_2 must have opposite signs. Thus, for a positive nonlinearity (e.g., the electronic third order nonlinearity in a glass fiber), this requires operation in the negative GVD region.

If the input intensity is slightly higher than needed for the fundamental soliton, then the pulse will shed some energy and eventually settle into the fundamental soliton. Thus, these pulses are very robust. At significantly higher intensities, higher order solitons can appear [49]. The input pulse will asymptotically approach the form of the n th order soliton, where n is the integer part of N , given by Eq. (91). The n th order solitons, for $n > 1$, change shape as they propagate, but recover to their original shape within a soliton period given

by

$$z_0 = \frac{\pi\tau_0^2}{2|k_2|} = 0.322 \frac{\pi t_{FWHM}^2}{2|k_2|} \quad (98)$$

The solitons described above are called bright solitons and have been observed experimentally [50]. They are the result of the interplay between self-phase modulation and anomalous group velocity dispersion. Interaction between self-phase modulation and *normal* group velocity dispersion ($k_2 > 0$) can lead to a phenomenon known as dark solitons. This is a pulse with a hole in the center that does not change shape as it propagates. The fundamental dark soliton takes the form $A(z, \tau) = A_0 \tanh(\tau/\tau_0) \exp(ikz)$. These have also been observed experimentally [51,52].

Spatial solitons. The nonlinear wave equation for a cw beam that varies rapidly in one space dimension (e.g., in a slab waveguide), such that its second order spatial derivative cannot be ignored, has the exact same form as the nonlinear Schrödinger equation introduced above. It follows that this wave equation admits the fundamental soliton solution given in Table 16, where w is the radius of the incident hyperbolic secant beam. This beam propagates in the nonlinear medium without spreading by diffraction (i.e., it is self-guided). This is reminiscent of the self-trapping described under Section III.E. However, the spatial soliton is stable with respect to minor perturbations.

Analogous to optical solitons (pulses) both bright and dark spatial solitons have been studied [53,54]. The generation of dark spatial solitons requires that a π -step phase or amplitude step be impressed on the input beam. The use of a spatial soliton to guide another beam has also been explored [55–57].

Early studies of the total reflection of a Gaussian beam from an interface between a linear medium and a nonlinear medium indicated that a spatial soliton could be propagated into the nonlinear medium for certain intensities [58]. These ideas have now been explored for propagation in a linear slab waveguide that is bounded on one side by a linear substrate and on the other side by a nonlinear cladding. For a positive nonlinearity, it has been determined that spatial solitons can be emitted into the nonlinear cladding where they propagate away from the waveguide [59–61]. Above a critical threshold intensity, the mode switches from a stable guided mode to a cladding mode. The soliton radiates away into the cladding, leaving a diminished guided mode in the waveguide. For increased incident intensity, there exist several thresholds for the emission of 1,2,3,... solitons, which shed energy from the guided mode.

Several applications based on soliton emission have been suggested [59]. These include optical limiting of the guided mode as excess energy is propagated into the cladding by the emitted solitons. Fast angular scanning of beams has also been proposed since the solitons propagate away from the waveguide at angles

that are intensity dependent. It also appears possible that coupling an emitted soliton into another nearby waveguide can form the basis for optical switches. Optical logic gates based on light intensity may then be constructed.

H. Nonlinear Optics in Fibers

Nonlinear optical effects are particularly efficient in optical fibers despite the relatively small nonlinear refractive index coefficient ($n_2 = 2.3 \times 10^{-22} \text{ m}^2/\text{V}^2$ or $n_2^I = 3.2 \times 10^{-16} \text{ cm}^2/\text{W}$ for silica fibers). The reason for this is that the spot size of radiation propagated in a single mode fiber is small ($\sim 2\text{--}4 \mu\text{m}$), this small spot size is maintained over a long interaction length by waveguiding, and the loss of optical fibers due to absorption and scattering can be quite small. The product IL_{eff} for a fiber can be $\sim 10^7\text{--}10^9$ times that in the same material in bulk form [49]. The primary reason is that for a correspondingly small spot size in the bulk material, diffraction severely limits the interaction length.

The phenomena discussed above (SPM, XPM, and solitons) are all quite efficient in optical fibers and have been studied extensively. Several other phenomena and applications have also been investigated. Agrawal [49] has devoted an entire book to the subject. Here two applications involving the nonlinear optics of ultrashort pulses in fibers are described.

Pulse compression. The principle of optical pulse compression is to pass a chirped pulse through a dispersive delay line. For a positive chirp pulse (i.e., blue-shifted frequencies in the trailing edge of the pulse), a negative GVD is required. As such a pulse travels in this medium, the red components in the leading edge of the pulse are delayed with respect to the blue components in the trailing edge. This slowing down of the front edge of the pulse compared to the end of the pulse causes the pulse to be compressed.

One of the most common pulse compressors is comprised of a fiber-grating pair. The fiber provides a positive, nearly linear chirp on the pulse through the combined effects of SPM and GVD. The diffraction grating pair provides an effective medium with negative GVD that is required to compress the positive chirp pulse. Light incident on the first grating is diffracted to the second grating, which then diffracts the light to the output. Because of the dispersive properties of the gratings, blue light travels a shorter distance and catches up with the red light, resulting in a compressed pulse. One drawback of this configuration is that due to the angular dispersion of wavelengths from a grating, the beam spreads spatially in one dimension. To overcome this, it is common to reflect the light from the second grating back through the compressor and pick off the compressed pulse with a beam splitter. This double-pass configuration mitigates the beam spreading and provides additional compression. The reflecting mirror is slightly tilted to avoid sending the pulse back through the fiber.

Fiber-grating pair compressors have yielded optical pulses with a width of a few femtoseconds. The detailed theory of their operation has been worked through numerical simulations. The objective of the compressor is to obtain a large compression factor in a high quality pulse, where nearly all of the pulse energy is maintained within the compressed pulse. This requires optimization of the fiber length to obtain the proper balance of SPM and GVD. Numerical simulations have shown that an optimum fiber length is given approximately by $(6L_{DNL})^{1/2}$ (i.e., approximately the geometric mean of the dispersion length and nonlinear length associated with the fiber).

Solitons in optical communications. One of the most important potential applications of optical solitons is high-speed data transmission in optical fibers. Short pulses that travel a long distance in fibers (several km) broaden due to group velocity dispersion and/or self-phase modulation. This limits the speed (bits/second) of data transmission since eventually the pulses spread into one another and can no longer be resolved. Solitons do not spread and hence can theoretically travel longer distances and/or have shorter widths for increased data transmission speed.

To preserve the soliton character of the pulses, a balance between the nonlinear and dispersive effects of the fiber must be maintained (i.e., the peak power of the pulses must be sustained). Therefore, optical loss in the fiber, although low, causes the width of the fundamental soliton to increase with the distance propagated. Numerical simulations show that over long distances ($\gg L_D$) the broadening is nearly linear with distance, though smaller than that in a linear medium. To provide data transmission over long distance (~ 1000 km), without the use of electronic repeaters, soliton schemes require repeater amplifiers to periodically boost the optical power and reshape the pulse.

Ideally, a pulse is launched in a soliton-based communication system with no chirp. In practice this is difficult to achieve. An initial chirp superimposed on the SPM induced chirp disturbs the balance between SPM and GVD needed for soliton propagation. Although solitons are stable for a weak initial chirp, some pulse energy is shed. Above a critical chirp value (dependent on the soliton order), the soliton can be destroyed.

The nonlinearity that provides the basis for soliton production also leads to interaction between solitons separated by small intervals of time. A soliton pair (separated by a time interval determined by the bit rate), launched with zero relative phase and equal amplitude, tend to attract one another. They will periodically collapse into a single soliton. If the relative phase of the pair is $\pi/4$, they will repel each other and become more separated in time as they propagate. Stable transmission can be achieved if soliton pairs are launched with unequal amplitudes.

The trend in fiber optic communications is to multiplex several channels at different wavelengths. This is known as wavelength division multiplexing

(WDM). To fully utilize the available bandwidth of the fiber, several densely spaced (in wavelength) signals will be propagated over a single fiber at the same time. Soliton propagation requires a small but finite dispersion. It is difficult to fabricate fibers that maintain this small dispersion value over the whole bandwidth of the fiber, however, it is not difficult to make a composite fiber with alternating sections of positive and negative GVD.

As a pulse enters the first section of such a fiber, it broadens and becomes chirped in one direction. Upon entering the second section, the opposite sign of the GVD produces an opposite chirp, causing the pulse to compress and become unchirped. Propagating further, the pulse once again broadens and becomes chirped in the opposite sense. It then enters the third fiber section where the opposite dispersion once again counteracts the effects of the previous section. In this manner, a stable soliton forms and “breathes” periodically, oscillating between large and narrow pulse width. The repeat length of the fibers of alternating GVD is usually the same as the spacing between erbium optical amplifiers—about 80 to 100 km.

These solitons are called dispersion-managed solitons. Dispersion-managed solitons are Gaussian shaped pulses whose shape is preserved as they propagate. Gaussian pulses have a much faster decay in the wings of the pulse than the hyperbolic-secant shaped pulses. There may thus be less interaction between neighboring soliton pairs, allowing the possibility of higher bit rates.

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