Nonlinear Optical Pulse Propagation

So far we have assumed that pulse propagation can be described by a linear transfer function

$$H(\omega) = R(\omega)\, e^{i\phi(\omega)}$$

Of course, ultrashort pulses with even a modest amount of energy have a high peak power. We shall find that under some circumstances, nonlinear effects modify the pulse propagation noticeably even for nanojoule pulses!

We will not undertake a systematic treatment of nonlinear optics, but will only consider the simplest case of a weak, nonresonant optical nonlinearity. (See Siegman 10.2 for a qualitative discussion.)

Recall that in a dielectric medium the linear response was described by a susceptibility

$$P(\omega) = \epsilon_0 \chi(\omega) E(\omega)$$

or, in the time domain,

$$P(t) = \epsilon_0 \int_{-\infty}^{t} \chi(t') E(t-t')\, dt'$$

so that $\chi(t)$ corresponds to the impulse response of the medium.

If the medium has a nonlinear response, we can loosely write

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

We could also write this as

$$P = P_L + P_{NL}$$

We will assume that $|P_{NL}| << |P_L|$.

For reasons of symmetry, the $\chi^{(2)}$ coefficient (2^{nd}-order nonlinear susceptibility) is nonzero only in crystals without inversion symmetry. The $\chi^{(2)}$ nonlinearity is responsible for second harmonic generation and parametric amplification; these are dealt with extensively in 538 and 634, so we will not consider these processes here.

In all materials, including isotropic materials (such as glass) and centrosymmetric crystals, $\chi^{(3)}$ is symmetry-allowed. It results from nonlinear distortions of the charge distribution in the material in response to the incident field.

In general

$$P^{(3)}(t) = \epsilon_0 \iiint dt_1 dt_2 dt_3 \, \chi^{(3)}(t_1, t_2, t_3) \cdot E(t-t_1) E(t-t_1-t_2) E(t-t_1-t_2-t_3)$$

so that the polarization may depend on the field at earlier times if the nonlinearity has "memory."

Note that:

(i) resonant $\chi^{(3)}$ interactions generally have "memory"; i.e. a ~~finite~~ nonzero response time, since atomic level populations are being changed ($10^{-8} - 10^{-11}$ s)

(ii) $\chi^{(3)}$ due to orientation of molecules can be large, but generally has a non-negligible response time (~ ps) due to the inertia of the molecules

(iii) nonresonant $\chi^{(3)}$ due to electronic nonlinear polarizability only has a "nearly instantaneous" (10^{-15} s) response time, but it's weak.

(see tables).

We shall begin by considering the case of a weak, nonresonant $\chi^{(3)}$ with instantaneous response.

This is essentially the case of a not-too-intense visible/near-IR pulse propagating in glass.

(Some data for different nonlinear processes is given on the following page.)

Assume pulses of form $E(t) = E(t)\cos\omega t$ (i.e. SVEA valid)

$$\Rightarrow \chi^{(3)}(t_1, t_2, t_3) = \chi^{(3)}(\omega_0)\,\delta(t_1)\,\delta(t_2)\,\delta(t_3)$$

$\chi^{(3)}(\omega_0)$: nonl. susceptibility constant over pulse bandwidth

$\delta(t_1)\,\delta(t_2)\,\delta(t_3)$: instantaneous response

TABLE 4.1.1 Typical values of the nonlinear refractive index*

Mechanism	n_2 (cm^2/W)	$\chi^{(3)}_{1111}$ (esu)	Response time (sec)
Electronic polarization	10^{-16}	10^{-14}	10^{-15}
Molecular orientation	10^{-14}	10^{-12}	10^{-12}
Electrostriction	10^{-14}	10^{-12}	10^{-9}
Saturated atomic absorption	10^{-10}	10^{-8}	10^{-8}
Thermal effects	10^{-6}	10^{-4}	10^{-3}
Photorefractive effect†	(large)	(large)	(intensity-dependent)

* For linearly polarized light, n_2 and $\chi^{(3)}$ are accurately related by Eq. (4.1.20).

† The photorefractive effect often leads to a very strong nonlinear response. These response usually cannot be described in terms of a $\chi^{(3)}$ (or an n_2) nonlinear susceptibility, because the nonlinear polarization does not depend on the applied field strength in the same manner as the other mechanisms listed.

TABLE 4.1.2 Third-order nonlinear susceptibilities of various materials*

Material	$\chi^{(3)}_{1111}$ (esu)	Response time
Air (20°C)	1.2×10^{-17}	
Carbon disulfide	1.9×10^{-12}	2 ps
GaAs (bulk, excitonic, room temperature)	6.5×10^{-4}	20 ns
GaAs/GaAlAs (MQW)	0.04	20 ns
Indium antimonide (77 K, 5.4 μm)	0.3	400 ns
Semiconductor-doped glass (containing CdSe)	10^{-8}	30 ps
Optical glasses	$(1\text{–}100) \times 10^{-14}$	Very fast
Polydiacetylene:		
Nonresonant	2.5×10^{-10}	Very fast
At peak of exciton	7.5×10^{-6}	2 ps

* The value of n_2 defined by $n = n_0 + n_2 I$ in units of cm^2/W can be obtained by multiplying the value of $\chi^{(3)}_{1111}$ by $0.0395/n_0^2$. The value of $\bar{n}_2$ defined by $n = n_0 + \bar{n}_2|E|^2$ in units of cm^3/erg can be obtained by multiplying the value of $\chi^{(3)}_{1111}$ by $3\pi/n_0$.

$$P^{(3)} = \epsilon_0 \chi^{(3)}(\omega) \left[\frac{1}{2} \left(E e^{i\omega t} + E^* e^{-i\omega t} \right) \right]^3$$

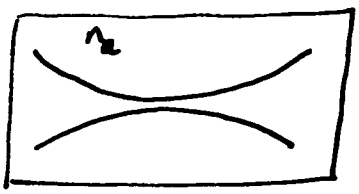
$$= \frac{\epsilon_0 \chi^{(3)}}{8} \left\{ \left[E^3 e^{i3\omega t} + \text{c.c.} \right] + \left[3|E|^2 E e^{i\omega t} + \text{c.c.} \right] \right\}$$

The first bracketed term describes third-harmonic generation. For this to be important, it must generally be phase-matched & is thus unimportant for propagation in bulk uniform media, so we will neglect it.

We are only interested in the component of the nonlinear polarization which is oscillating at frequency ω (i.e. the same frequency as the input field).

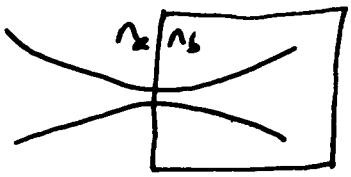
Note that third harmonic generation can be significant at an interface when the beam is tightly focussed

in bulk:



each: 180° Gouy phase shift
⇒ 3ω signal from left of focus is 180° out of phase w.r.t. 3ω from right of focus
⇒ no net 3ω generation!

at interface



- 3ω from left of focus is no longer 180° out of phase w.r.t 3ω from right ⇒ nonzero net 3ω signal

We will neglect the third-harmonic generation process here, and assume a nonresonant mechanism so $\chi^{(3)}$ is <u>real</u> (so induced polarization is real).

$$P = \epsilon_0 \chi^{(1)} \mathcal{E} + \frac{3}{4} \epsilon_0 \chi^{(3)} |\mathcal{E}|^2 \mathcal{E}$$

Recall $D = \epsilon_0 \mathcal{E} + P \equiv \epsilon \mathcal{E}$

$$= \epsilon_0 \left[1 + \chi^{(1)} + \frac{3}{4} \chi^{(3)} |\mathcal{E}|^2 \right] \mathcal{E}$$

$$\Rightarrow \epsilon = n^2 = \epsilon_0 \left[1 + \chi^{(1)} + \frac{3}{4} \chi^{(3)} |\mathcal{E}|^2 \right]$$

$$= n_0^2 \left[1 + \frac{3}{4 n_0^2} \chi^{(3)} |\mathcal{E}|^2 \right]$$

$\Rightarrow$ For a small nonlinearity, the index of refraction is approximately

$$n = n_0 + \frac{3 \chi^{(3)}}{8 n_0} |\mathcal{E}|^2$$

or $\boxed{n = n_0 + n_2 |\mathcal{E}|^2}$ where $n_2 = \frac{3 \chi^{(3)}}{8 n_0}$

This is frequently written as

$\boxed{n = n_0 + \bar{n}_2 I}$ where $\bar{n}_2 = \frac{2 n_2}{\epsilon_0 c n_0}$

units: $\bar{n}_2 = \frac{cm^2}{W}$ usually (since $I \sim \frac{W}{cm^2}$)

Frequently $\chi^{(3)}$ is tabulated in esu units. In that case, it can be shown that

$$\bar{n}_2 \left(\frac{cm^2}{W}\right) = \frac{12\pi^2}{n_0^2 c} 10^7 \chi^{(3)} \text{ (esu)}$$

$$= \frac{0.0395}{n_0^2} \chi^{(3)} \text{ (esu)}$$

Some typical numbers are given on p. 165.5.

Note that generally $\underline{n_2 > 0}$.

Self-Phase Modulation

Before going on to consider nonlinear propagation in dispersive media, let's consider the simplest consequence of a nonzero n_2: the conversion of amplitude modulation into phase modulation.

Suppose a wavepacket propagates through a medium which is short compared to a dispersion length.

$$\mathcal{E}(z,t) = \mathbf{E}(t) e^{i\phi(z,t)}$$

$$\phi(t) = \omega_0 t - \beta z \qquad \beta = \frac{n\omega_0}{c}$$

$$\varphi(t) = \omega_0 t - \frac{\omega_0}{c}\left[n_0 + \bar{n}_2 I(t)\right] z$$

$$= \omega_0 t - \beta_0 z - \frac{\bar{n}_2 \omega_0}{c} z\, I(t) \qquad \beta_0 = \frac{n_0 \omega_0}{c}$$

$$= \omega_0 t - \beta_0 z + \varphi_{NL}(z,t)$$

where $\varphi_{NL} = -\frac{\bar{n}_2 \omega_0}{c} z\, I(t)$

Recall we defined the instantaneous frequency as

$$\omega = \frac{d\varphi}{dt} = \omega_0 - \frac{\bar{n}_2 \omega_0}{c} z \frac{dI(t)}{dt}$$

$\Rightarrow$ the nonlinearity induces a frequency shift

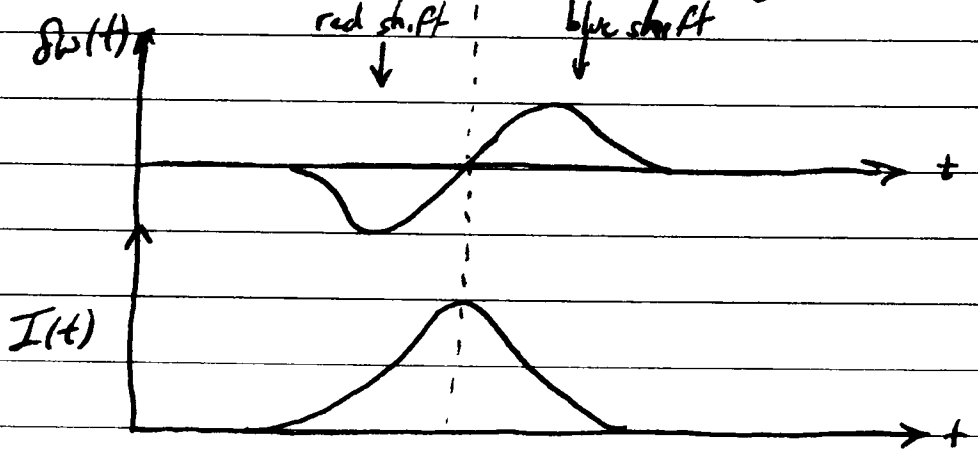
$$\boxed{\delta\omega = -\frac{\bar{n}_2 \omega_0}{c} z \frac{dI}{dt}}$$

Thus the effects of the nonlinearity are

(1) to broaden the pulse spectrum (add new frequencies)

(2) to induce a chirp on the pulse

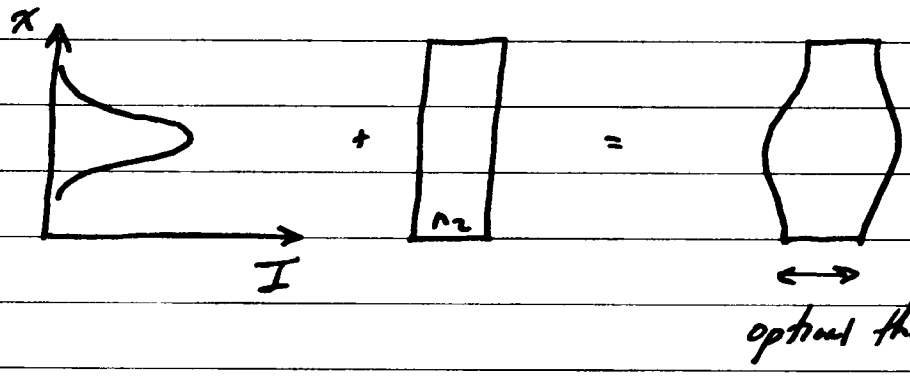
(remember we're still ignoring dispersion)



Self-focussing

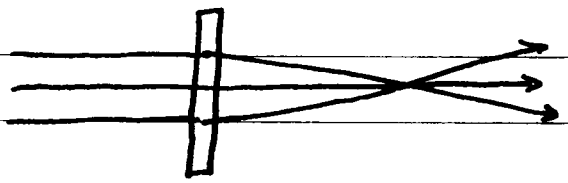
A second physical consequence of $n_2 > 0$ is the phenomenon of self-focussing.

$n = n_0 + \bar{n}_2 I$ ⇒ n is larger at the center of a Gaussian beam than at the edges



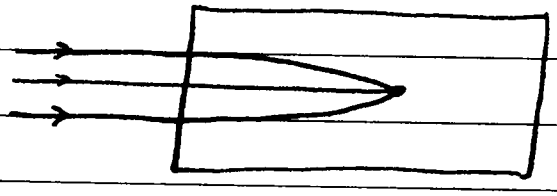
Thus a plane-parallel slab of glass becomes equivalent to a positive lens in the nonlinear regime. If the intensity is high enough, this positive lens will be large enough to overcome the natural divergence of a beam due to diffraction, and the beam will focus itself.

short medium:



(note: this is the basis of the "Z-scan" technique, which is an accurate method of measuring n_2)

long medium :



Note that, as the beam focusses, the intensity increases, resulting in a stronger lens effect, etc. The process essentially runs away to a catastrophic self-focussing leading to breakdown (damage) of the material.

It is clear that a full treatment of ultrashort pulses in n_2 media is a complicated 4-D problem (i.e. 3 spatial dimensions plus time, since the nonlinearity couples the spatial and temporal behavior)!
In other words, in bulk dielectrics, non-negligible SPM is always accompanied by non-negligible self-focussing, so it's not really legitimate for us to consider 1-D approximate models.

In single-mode optical fibers, however, the transverse profile of the beam is defined by the fundamental mode of the fiber, and self-focussing does not occur. Thus we have effectively a 1-D propagation problem, and we can consider the temporal behavior of a complex field envelope as a function of propagation distance.