
Nonlinear Optics

OPTICS WITH $\chi^{(3)}$ MATERIALS

0.1 Introduction

The main idea is still to use the propagation equation derived from the Maxwell's equation

$$\frac{\partial^2 E}{\partial z^2} - \frac{n^2}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2}{\partial t^2} P_{\text{NL}} \quad (1)$$

and which does not depend on the form of the nonlinear polarization P_{NL} . By contrast with non-centrosymmetric crystal all material exhibit third order susceptibility and it is therefore important to study - at least - a few of the nonlinear effects that can result from it.

0.1.1 Nonlinear refractive index

In this case we assume a monochromatic input wave

$$E = \mathcal{E} e^{i\omega t} + c.c. \quad (2)$$

The nonlinear polarization $P_{\text{NL}} = \epsilon \chi^{(3)} E^3$ can be readily calculated

$$P_{\text{NL}} = \epsilon_0 \chi^{(3)} (3 |\mathcal{E}|^2 \mathcal{E} e^{i\omega t} + c.c. + \mathcal{E} e^{3i\omega t} + c.c.) \quad (3)$$

From this nonlinear polarization we see that two processes are possible:

1. The input field creates a modification of the refractive index

$$n = n_0 + n_2 |\mathcal{E}|^2 \quad (4)$$

where n_0 is the linear refractive index, and n_2 is the nonlinear refractive index coefficient.

2. The input field yields a polarization oscillating at 3ω and this will act as a source term in the propagation equation. It then includes solutions oscillating at this frequency, which is three times larger than the pump frequency: this is the phenomenon of *third harmonic generation*.

0.1.2 Multiple wave mixing

Things become complicated and challenging and more interesting when multiple frequencies interact in the nonlinear material. Let's assume that we have an input consisting of three waves¹ interacting inside our third order material

$$E = E_1(\omega_1) + E_2(\omega_2) + E_3(\omega_3) \quad (5a)$$

$$\text{with } E_j(\omega_j) = \mathcal{E}_j(\omega_j, z, t) e^{i\omega_j t} + c.c \quad (5b)$$

¹Note that in principle we can have up to 4 waves interacting inside the crystal through a third-order nonlinearity.

In this case the nonlinear polarization is

$$P_{\text{NL}} = P_{\text{NL}}(3\omega) + \sum_{j=1}^3 P_{\text{NL},j} \quad (6)$$

with

$$P_{\text{NL}}(3\omega) = \epsilon_0 \chi^{(3)} \sum_{k=1}^3 \mathcal{E}_k^3 e^{3i\omega_k t} + c.c. \quad (7a)$$

$$\begin{aligned} P_{\text{NL},1} &= 3\epsilon_0 \chi^{(3)} (|\mathcal{E}_1|^2 + 2|\mathcal{E}_2|^2 + 2|\mathcal{E}_3|^2) \mathcal{E}_1 e^{i\omega_1 t} \\ &+ 3\epsilon_0 \chi^{(3)} (2|\mathcal{E}_1|^2 + |\mathcal{E}_2|^2 + 2|\mathcal{E}_3|^2) \mathcal{E}_2 e^{i\omega_2 t} \\ &+ 3\epsilon_0 \chi^{(3)} (2|\mathcal{E}_1|^2 + 2|\mathcal{E}_2|^2 + |\mathcal{E}_3|^2) \mathcal{E}_3 e^{i\omega_3 t} + c.c. \end{aligned} \quad (7b)$$

$$\begin{aligned} \frac{P_{\text{NL},2}}{\epsilon_0 \chi^{(3)}} &= 3\mathcal{E}_1^2 \mathcal{E}_2 e^{i(2\omega_1 + \omega_2)t} + 3\mathcal{E}_1^2 \mathcal{E}_3 e^{i(2\omega_1 + \omega_3)t} \\ &+ 3\mathcal{E}_1^2 \mathcal{E}_2^* e^{i(2\omega_1 - \omega_2)t} + 3\mathcal{E}_1^2 \mathcal{E}_3^* e^{i(2\omega_1 - \omega_3)t} \\ &+ 3\mathcal{E}_2^2 \mathcal{E}_1 e^{i(2\omega_2 + \omega_1)t} + 3\mathcal{E}_2^2 \mathcal{E}_3 e^{i(2\omega_2 + \omega_3)t} \\ &+ 3\mathcal{E}_2^2 \mathcal{E}_1^* e^{i(2\omega_2 - \omega_1)t} + 3\mathcal{E}_2^2 \mathcal{E}_3^* e^{i(2\omega_2 - \omega_3)t} \\ &+ 3\mathcal{E}_3^2 \mathcal{E}_1 e^{i(2\omega_3 + \omega_1)t} + 3\mathcal{E}_3^2 \mathcal{E}_2 e^{i(2\omega_3 + \omega_2)t} \\ &+ 3\mathcal{E}_3^2 \mathcal{E}_1^* e^{i(2\omega_3 - \omega_1)t} + 3\mathcal{E}_3^2 \mathcal{E}_2^* e^{i(2\omega_3 - \omega_2)t} + c.c. \end{aligned} \quad (7c)$$

$$\begin{aligned} \frac{P_{\text{NL},3}}{\epsilon_0 \chi^{(3)}} &= 6\mathcal{E}_1 \mathcal{E}_2 \mathcal{E}_3 e^{i(\omega_1 + \omega_2 + \omega_3)t} + 6\mathcal{E}_1 \mathcal{E}_2^* \mathcal{E}_3 e^{i(\omega_1 - \omega_2 + \omega_3)t} \\ &+ 6\mathcal{E}_1 \mathcal{E}_2 \mathcal{E}_3^* e^{i(\omega_1 + \omega_2 - \omega_3)t} + 6\mathcal{E}_1 \mathcal{E}_2^* \mathcal{E}_3^* e^{i(\omega_1 - \omega_2 - \omega_3)t} + c.c. \end{aligned} \quad (7d)$$

This is obviously more complicated than in the case of a monochromatic wave. We can however quite straightforwardly identify that the first term (eq. (7a)) is a generalization of the third harmonic generation that we already saw in the case of a monochromatic input (eq. (3)). On the other hand the second term $P_{\text{NL},1}$, which is the nonlinear modification of the refractive index is much less obvious. Indeed we see that the contribution oscillating at $\exp i\omega_j t$ (with $j = 1, 2, 3$) includes term as $3(|\mathcal{E}_j|^2 + 2|\mathcal{E}_k|^2 + 2|\mathcal{E}_l|^2) \mathcal{E}_j$. This clearly contrast with the simpler term $3|\mathcal{E}_j|^2 \mathcal{E}_j$ that we obtained in the case of a monochromatic input.

These more complicated contributions are actually a manifestation of the interference between two distinct fields \mathcal{E}_j and $\mathcal{E}_{k,l}$. As a general observation these *cross-terms* between fields are affected by a factor 2. As we will see later the term $3|\mathcal{E}_j|^2 \mathcal{E}_j$ leads to the so-called self-phase modulation phenomena while the more complicated terms yield *cross-phase* modulation.

The last two terms represent the possible interaction between two and three waves and are direct generalizations of the sum and difference frequencies processes as we saw them in the chapter related to $\chi^{(2)}$ crystals.

0.2 Optical phase-conjugation mirror

0.2.1 Linear case: Fresnel's law

It is well known that when a beam is incident on a surface separating two media (Fig.) it is affected in such a way that part of the beam is reflected and remains in the same

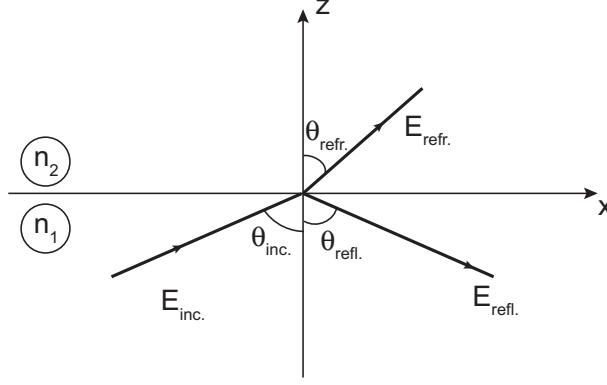


Figure 1: Refraction and reflection of a beam at an interface between two media. The refractive indices are $n_1 < n_2$.

medium (refractive index n_1) whilst the another part is refracted into the second medium (refractive index n_2)

The incident field (resp. reflected and refracted ones) can be defined as

$$E_{\text{inc.}} = A \exp [i (\mathbf{k} \cdot \mathbf{r} - \omega t)] + c.c. \quad (8a)$$

$$E_{\text{refr.}} = A' \exp [i (\mathbf{k}' \cdot \mathbf{r} - \omega t)] + c.c. \quad (8b)$$

$$E_{\text{refl.}} = A'' \exp [i (\mathbf{k}'' \cdot \mathbf{r} - \omega t)] + c.c. \quad (8c)$$

where A is the amplitude of the incident field and \mathbf{k} its wave-vector, which has a norm $k = (n\omega/c)$ where ω is the angular frequency of the wave. Considering the boundaries conditions at the interface we can simply write that $k \sin \theta_{\text{inc}} = k' \sin \theta_{\text{refr.}} = k'' \sin \theta_{\text{refl.}}$. And since $k \equiv k''$ then $\theta_{\text{inc.}} = \theta_{\text{refl.}}$. The other relation yields the well-known Snell's law

$$\frac{\sin \theta_{\text{inc}}}{\sin \theta_{\text{refr.}}} = \frac{k'}{k} = \frac{n'}{n} \quad (9)$$

Although the Snell's laws lead to the direction of the reflected and refracted beam they do not give any indication of the amplitude of the reflected and refracted beam compare to the incident one. That is precisely what the Fresnel coefficients give. Without entering the details of the derivation of these laws² we can simply give the main results:

$$A'' = r \cdot A \quad (10a)$$

$$A' = t \cdot A \quad (10b)$$

$$(10c)$$

where the coefficient r and t are respectively the reflection and the transmission coefficient. The conservation of energy imposes that $r^2 + t^2 = 1$.

0.2.2 Nonlinear case

the problem of optical phase conjugation mirror

In the nonlinear case, we replace the traditional mirror by a $\chi^{(3)}$ material. For a very specific set of parameters, which we will establish in the following, this can create a very peculiar mirror where an input field E_{probe} is reflected by the $\chi^{(3)}$ material (Fig. 2). More surprising the reflected field is the conjugate of the input one: this is an *optical phase conjugation* setup (OPC).

²the coefficient are also dependent of the polarization of the wave, but we do not need to consider this here.

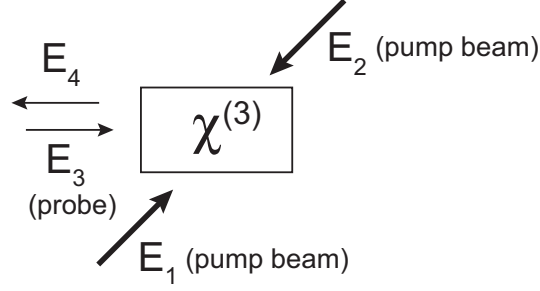


Figure 2: Schematic of an optical phase conjugation mirror.

As shown on the Fig. 2, to obtain an OPC we need

1. E_1 and E_2 are the pump beam. Their frequencies are ω_1 and ω_2 resp. and they are contra-propagating. Since their wave-vectors are resp. \mathbf{k}_1 and \mathbf{k}_2 we can say that

$$\frac{\mathbf{k}_1}{|\mathbf{k}_1|} + \frac{\mathbf{k}_2}{|\mathbf{k}_2|} = 0 \quad (11)$$

The interaction between these fields creates a grating through their interaction in the nonlinear material.

2. The probe beam (E_3) is sent onto the nonlinear material. Its frequency is ω_3 and its wave-vector \mathbf{k}_3 . It will interact with the grating generated by E_1 & E_2 and eventually be reflected.
3. The fourth wave interacting in the crystal is E_4 . Since this is – hopefully – the reflection of E_3 , the direction of propagation of E_4 is opposite to the one of E_3 . Therefore we have

$$\frac{\mathbf{k}_3}{|\mathbf{k}_3|} + \frac{\mathbf{k}_4}{|\mathbf{k}_4|} = 0 \quad (12)$$

As we already mentioned we will see that E_4 may have very special properties such as optical phase conjugation

$$E_3 = A_3 e^{-i(\omega_3 t - \mathbf{k}_3 \cdot \mathbf{r})} + c.c \implies E_4 = A_3^* e^{-i(\omega_4 t - \mathbf{k}_4 \cdot \mathbf{r})} + c.c \quad (13)$$

The remaining question is *How to get such property?*

Conditions for optical phase conjugation

As before, we need to calculate the nonlinear polarization $P_{NL} = \epsilon_0 \chi^{(3)} E^3$ resulting from the interaction between the four fields

$$E = \sum_{j=1}^4 A_j e^{-i(\omega_j t - \mathbf{k}_j \cdot \mathbf{r})} \quad (14)$$

Since we do not want to calculate all the terms from the nonlinear polarization we will only consider the case where

$$\begin{cases} \omega_1 + \omega_2 = \omega_3 + \omega_4 \\ \mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3 + \mathbf{k}_4 = 0 \text{ since } E_1 \text{ and } E_2 \text{ have opposite direction} \end{cases} \quad (15)$$

The different contribution to the polarization are

$$\frac{P_1}{\epsilon_0\chi^{(3)}} = 3 \left[(|A_1|^2 + 2|A_2|^2 + 2|A_3|^2 + 2|A_4|^2) A_1 + 2A_2^*A_3A_4 \right] e^{-i(\omega_1 t - \mathbf{k}_1 \cdot \mathbf{r})} \quad (16a)$$

$$\frac{P_2}{\epsilon_0\chi^{(3)}} = 3 \left[(2|A_1|^2 + |A_2|^2 + 2|A_3|^2 + 2|A_4|^2) A_2 + 2A_1^*A_3A_4 \right] e^{-i(\omega_2 t - \mathbf{k}_2 \cdot \mathbf{r})} \quad (16b)$$

$$\frac{P_3}{\epsilon_0\chi^{(3)}} = 3 \left[(2|A_1|^2 + 2|A_2|^2 + |A_3|^2 + 2|A_4|^2) A_3 + 2A_1A_2A_4^* \right] e^{-i(\omega_3 t - \mathbf{k}_3 \cdot \mathbf{r})} \quad (16c)$$

$$\frac{P_4}{\epsilon_0\chi^{(3)}} = 3 \left[(2|A_1|^2 + 2|A_2|^2 + 2|A_3|^2 + |A_4|^2) A_4 + 2A_1A_2A_3^* \right] e^{-i(\omega_4 t - \mathbf{k}_4 \cdot \mathbf{r})} \quad (16d)$$

Now, let us assume that the so-called *pump*-beam E_1 and E_2 are much more intense than the two other fields

$$|E_1|, |E_2| \gg |E_3|, |E_4| \quad (17)$$

We can then consider that they are not affected by the presence of E_3 and E_4 . Using this assumption we can reduce the eq. (16) to

$$\frac{P_1}{\epsilon_0\chi^{(3)}} = 3 (|A_1|^2 + 2|A_2|^2) A_1 e^{-i(\omega_1 t - \mathbf{k}_1 \cdot \mathbf{r})} \quad (18a)$$

$$\frac{P_2}{\epsilon_0\chi^{(3)}} = 3 (2|A_1|^2 + |A_2|^2) A_2 e^{-i(\omega_2 t - \mathbf{k}_2 \cdot \mathbf{r})} \quad (18b)$$

$$\frac{P_3}{\epsilon_0\chi^{(3)}} = 6 \{ (|A_1|^2 + |A_2|^2) A_3 + A_1A_2A_4^* \} e^{-i(\omega_3 t - \mathbf{k}_3 \cdot \mathbf{r})} \quad (18c)$$

$$\frac{P_4}{\epsilon_0\chi^{(3)}} = 6 \{ (|A_1|^2 + |A_2|^2) A_4 + A_1A_2A_3^* \} e^{-i(\omega_4 t - \mathbf{k}_4 \cdot \mathbf{r})} \quad (18d)$$

We now need to use these nonlinear contributions in the propagation equation (eq. (1)). Using the slow-varying envelope approximation and since the system is *free-running* we obtain after calculations

$$\frac{dA_1}{dz'} = \frac{3i\omega_1\chi^{(3)}}{2n_1\epsilon_0c} (|A_1|^2 + 2|A_2|^2) A_1 \quad (19a)$$

$$\frac{dA_2}{dz'} = \frac{-3i\omega_2\chi^{(3)}}{2n_2\epsilon_0c} (2|A_1|^2 + |A_2|^2) A_2 \quad (19b)$$

$$(19c)$$

where the $-$ sign comes from the fact that we are using direction of propagation of E_1 (z') for the derivation and E_1 and E_2 have opposite direction. Note that as we noticed in the case of second harmonic we have here

$$\frac{d|A_1|^2}{dz'} = \frac{d|A_2|^2}{dz'} = 0 \quad (20)$$

meaning that the pump beams E_1 and E_2 are not affected during the process. This also means that the pump beams simply acquire a phase-shift during their propagation in the third-order material

$$A_1(z') = A_1(0) e^{i\gamma_1 (|A_1|^2 + 2|A_2|^2) z'} \quad (21a)$$

$$A_2(z') = A_2(0) e^{-i\gamma_2 (2|A_1|^2 + |A_2|^2) z'} \quad (21b)$$

where $\gamma_i = (3\omega_j\chi^{(3)}/2n_j\epsilon_0c)$. Note that in the particular case where $\gamma_1 (|A_1|^2 + 2|A_2|^2) = \gamma_2 (2|A_1|^2 + |A_2|^2)$, which is equivalent as

$$\begin{aligned} \left(\frac{3\omega_1\chi^{(3)}}{2n_1\epsilon_0c}\right) (|A_1|^2 + 2|A_2|^2) &= \left(\frac{3\omega_2\chi^{(3)}}{2n_2\epsilon_0c}\right) (2|A_1|^2 + |A_2|^2) \\ \Leftrightarrow n_2\omega_1 (|A_1|^2 + 2|A_2|^2) &= n_1\omega_2 (2|A_1|^2 + |A_2|^2) \\ \Leftrightarrow \frac{|A_1|^2}{|A_2|^2} &= \frac{2n_2\omega_1 - n_1\omega_2}{2n_1\omega_2 - n_2\omega_2} \end{aligned} \quad (22)$$

then the product of the field is space-invariant:

$$A_1(z')A_2(z') = A_1(0)A_2(0) \quad (23)$$

Similarly the coupled equations for the probe beam are

$$\frac{dA_3}{dz} = \frac{3i\omega_3\chi^{(3)}}{2n_3\epsilon_0c} \{(|A_1|^2 + |A_2|^2) A_3 + A_1A_2A_4^*\} \quad (24a)$$

$$\frac{dA_4}{dz} = \frac{-3i\omega_4\chi^{(3)}}{2n_4\epsilon_0c} \{(|A_1|^2 + |A_2|^2) A_4 + A_1A_2A_3^*\} \quad (24b)$$

$$(24c)$$

In the particular case where the assumption (eq. (22)) is fulfilled then the coefficients of $(|A_1|^2 + |A_2|^2)$ as well as A_1A_2 are space-invariant and therefore the coupled equations (eq. (24)) are linear. Additionally in the case where $\omega_3/n_3 = \omega_4/n_4$ then these equations (eq. (24)) have a phase conjugation solution

$$A_4(z) = A_3^*(z) \quad (25)$$

If we have $\omega_3/n_3 \neq \omega_4/n_4$ then the general solution of the coupled equations (eq. (24)) is

$$A_4(z) = \frac{n_3\omega_4}{n_4\omega_3} A_3^*(z) \quad (26)$$

This is quite surprising since the *reflection coefficient* $r = (n_3\omega_4/n_4\omega_3)$ can be larger than unity. Not only the input beam is reflected and possesses the properties of phase-conjugation but it can also be amplified!

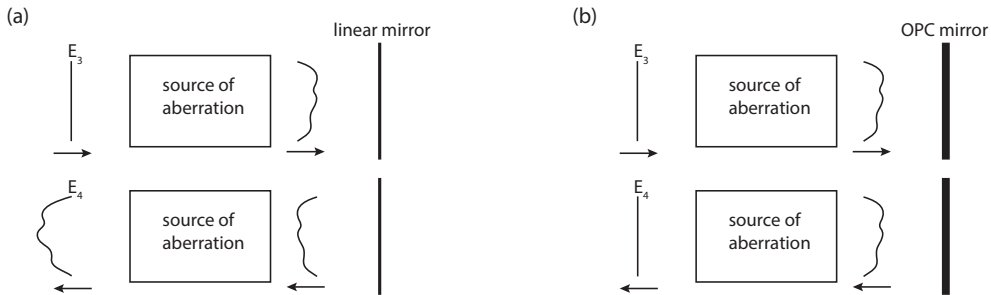


Figure 3: Comparison between (a) a linear mirror and (b) a optical phase conjugation mirror for compensation of phase aberration.

Obviously these results require many specific conditions but it contrasts so much with a traditional mirror that it is an important application of third-order nonlinearity. If we remind that in the case of a linear mirror we had $A_4(z) = rA_3(z)$ with $r < 1$ we

can clearly see that the situation is totally different here. In particular since the pump beam are contra-propagating ($\mathbf{k}_1 + \mathbf{k}_2 = 0$) then the wave-vector of the reflected beam \mathbf{k}_4 has to fulfill ($\mathbf{k}_3 + \mathbf{k}_4 = 0$). This means $(k_{4x}, k_{4y}, k_{4z}) = (-k_{3x}, -k_{3y}, -k_{3z})$. All three components³ are reversed! It is particularly important to balance the effect of phase aberration, which could yield modification of the wavefront of a beam (Fig. 3).

³This is obviously in strong contrast with the case of a linear mirror where we have $(k_{4x}, k_{4y}, k_{4z}) = (k_{3x}, k_{3y}, -k_{3z})$.