Chapter 2

Wave-Equation Description of Nonlinear Optical Interactions

Spring 2021 Non-linear optics study

Nonlinear Optics, 4th edition, Robert W. Boyd
Chapter 1

The Nonlinear Optical Susceptibility

Quick Recall: Nonlinear optical effects and models, including:

- Second Harmonic Generation
- Sum and Difference Generation
- Third Order Nonlinearities
- Parametric and Non-parametric Processes
- Classical Nonlinear Optical Model
- Symmetries of Optical Tensors
Developing the Nonlinear Optical Wave Equation

2.1 The Wave Equation for Nonlinear Optical Media

We have seen in the last chapter how nonlinearity in the response of a material system to an intense laser field can cause the polarization of the medium to develop new frequency components not present in the incident radiation field. These new frequency components of the nonlinear polarization act as sources of new frequency components of the electromagnetic field. In the present chapter, we examine how Maxwell’s equations describe the generation of these new components, and more generally we see how the various frequency components of the field become coupled by the nonlinear interaction.
Physical Picture of Nonlinear Frequency Generation

(a) \( \omega_1 \) \( \omega_2 \) \( \omega_1 + \omega_2 \) \( \omega_3 = \omega_1 + \omega_2 \)

(b) \( \omega_1 + \omega_2 \)

(c) \( \omega_1 + \omega_2 \)
Developing the Nonlinear Optical Wave Equation

“The system will act as a phased array of dipoles when a certain condition, known as the phase-matching condition…[Section 2.2]…is satisfied.”
Developing the Nonlinear Optical Wave Equation

Maxwell’s Equations:

\[ \nabla \cdot \vec{D} = \rho, \quad (2.1.1) \]
\[ \nabla \cdot \vec{B} = 0, \quad (2.1.2) \quad \text{Gauss’s Law for Magnetism} \]
\[ \nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t}, \quad (2.1.3) \quad \text{Faraday’s Law} \]
\[ \nabla \times \vec{H} = \frac{\partial \vec{D}}{\partial t} + \vec{J}. \quad (2.1.4) \quad \text{Ampere’s Law} \]
Developing the Nonlinear Optical Wave Equation

Maxwell’s Equations: Initial assumptions:

\[ \tilde{\rho} = 0, \quad (2.1.5) \]

\[ \tilde{\mathbf{J}} = 0. \quad (2.1.6) \]

\[ \tilde{\mathbf{B}} = \mu_0 \tilde{\mathbf{H}}. \quad (2.1.7) \]

\[ \tilde{\mathbf{D}} = \varepsilon_0 \tilde{\mathbf{E}} + \tilde{\mathbf{P}}, \quad (2.1.8) \]
Developing the Nonlinear Optical Wave Equation

We now proceed to derive the optical wave equation in the usual manner. We take the curl of the curl-$\mathbf{\tilde{E}}$ Maxwell equation (2.1.3), interchange the order of space and time derivatives on the right-hand side of the resulting equation, and use Eqs. (2.1.4), (2.1.6), and (2.1.7) to replace $\nabla \times \mathbf{\tilde{B}}$ by $\mu_0 (\partial \mathbf{\tilde{D}} / \partial t)$, to obtain the equation

$$\nabla \times \nabla \times \mathbf{\tilde{E}} + \mu_0 \frac{\partial^2 \mathbf{\tilde{D}}}{\partial t^2} = 0.$$  \hspace{1cm} (2.1.9a)

We now use Eq. (2.1.8) to eliminate $\mathbf{\tilde{D}}$ from this equation, and we thereby obtain the expression

$$\nabla \times \nabla \times \mathbf{\tilde{E}} + \frac{1}{c^2} \frac{\partial^2 \mathbf{\tilde{E}}}{\partial t^2} = - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \mathbf{\tilde{P}}}{\partial t^2}.$$ \hspace{1cm} (2.1.9b)
Developing the Nonlinear Optical Wave Equation

\[ \nabla \times \nabla \times \tilde{E} + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \tilde{E} = - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \tilde{P}}{\partial t^2}. \] (2.1.9b)

This is the most general form of the wave equation in nonlinear optics. Under certain conditions it can be simplified. For example, by using an identity from vector calculus, we can write the first term on the left-hand side of Eq. (2.1.9b) as

\[ \nabla \times \nabla \times \tilde{E} = \nabla (\nabla \cdot \tilde{E}) - \nabla^2 \tilde{E}. \] (2.1.10)

The first term on the right side of this equation goes to zero or becomes small when the incident electric field is a transverse, infinite plane wave, or when we are using the slowly varying amplitude approximation, as shown in the next section.

\[ \nabla \times \nabla \times \tilde{E} = - \nabla^2 \tilde{E}. \]
Developing the Nonlinear Optical Wave Equation

For the remainder of this book, we shall usually assume that the contribution of $\nabla(\nabla \cdot \mathbf{E})$ in Eq. (2.1.10) is negligible so that the wave equation can be taken to have the form

$$
\nabla^2 \mathbf{E} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E} = \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \mathbf{P}}{\partial t^2}.
$$

(2.1.11)

Alternatively, the wave equation can be expressed as

$$
\nabla^2 \mathbf{E} - \frac{1}{\varepsilon_0 c^2} \frac{\partial^2}{\partial t^2} \mathbf{D} = 0
$$

(2.1.12)
Developing the Nonlinear Optical Wave Equation

For this (simpler) case of an isotropic, dispersionless material, the wave equation (2.1.15) becomes

\[
\nabla^2 \tilde{E} - \frac{\epsilon^{(1)}}{c^2} \frac{\partial^2 \tilde{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \tilde{P}_{NL}^n}{\partial t^2}.
\]  

(2.1.17)

For the case of a dispersive medium,

\[
\nabla^2 \tilde{E}_n - \frac{\epsilon^{(1)}(\omega_n)}{c^2} \frac{\partial^2 \tilde{E}_n}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \tilde{P}_{NL}^n}{\partial t^2}.
\]

(2.1.21)
2.2 The Coupled-Wave Equations for Sum-Frequency Generation

We next study how the nonlinear optical wave equation that we derived in the previous section can be used to describe specific nonlinear optical interactions. In particular, we consider sum-frequency generation in a lossless nonlinear optical medium involving collimated, monochromatic, continuous-wave input beams. We assume the configuration shown in Fig. 2.2.1.
Coupled Wave Equations (for SFG)

The wave equation in Eq. (2.1.21) must hold for each frequency component of the field and in particular for the sum-frequency component at frequency $\omega_3$. In the absence of a nonlinear source term, the solution to this equation for a plane wave at frequency $\omega_3$ propagating in the $+z$ direction is

$$\tilde{E}_3(z, t) = A_3 e^{i(k_3 z - \omega_3 t)} + c.c., \quad (2.2.1)$$

where\(^\dagger\)

$$k_3 = \frac{n_3 \omega_3}{c}, \quad n_3^2 = \varepsilon^{(1)}(\omega_3), \quad (2.2.2)$$

and where the amplitude of the wave $A_3$ is a constant.
Coupled Wave Equations (for SFG)

We expect on physical grounds that, when the nonlinear source term is not too large, the solution to Eq. (2.1.21) will still be of the form of Eq. (2.2.1), except that $A_3$ will become a slowly varying function of $z$. We hence adopt Eq. (2.2.1) with $A_3$ taken to be a function of $z$ as the form of the trial solution to the wave equation (2.1.21) in the presence of the nonlinear source term.

$$\tilde{E}_3(z, t) = A_3 e^{i(k_3z - \omega_3 t)} + \text{c.c.}, \quad (2.2.1)$$
Coupled Wave Equations (for SFG)

Defining the terms of our model:

\[
\tilde{E}_3(z, t) = A_3 e^{i(k_3 z - \omega_3 t)} + \text{c.c.,} \quad (2.2.1)
\]

\[
\tilde{P}_3(z, t) = P_3 e^{-i\omega_3 t} + \text{c.c.,} \quad (2.2.3)
\]

\[
P_3 = 4\varepsilon_0 d_{\text{eff}} E_1 E_2. \quad (2.2.4)
\]

We represent the applied fields \((i = 1, 2)\) as

\[
\tilde{E}_i(z, t) = E_i e^{-i\omega_i t} + \text{c.c.,} \quad \text{where} \quad E_i = A_i e^{i k_i z}. \quad (2.2.5)
\]

The amplitude of the nonlinear polarization can then be written as

\[
P_3 = 4\varepsilon_0 d_{\text{eff}} A_1 A_2 e^{i(k_1 + k_2) z} \equiv p_3 e^{i(k_1 + k_2) z}. \quad (2.2.6)
\]
Coupled Wave Equations (for SFG)

Plugging all of these terms into our nonlinear optical wave equation, eq. 2.1.21:

\[
\frac{d^2 A_3}{dz^2} + 2ik_3 \frac{dA_3}{dz} = \frac{-4d_{\text{eff}}\omega_3^2}{c^2} A_1 A_2 e^{i(k_1+k_2-k_3)z}. \tag{2.2.8}
\]

Using the slowly varying amplitude approximation (very small changes in $A_3$ over a length in $z$ of one wavelength), we conclude that the first term is negligible with respect to the second term:

\[
\frac{dA_3}{dz} = \frac{2id_{\text{eff}}\omega_3}{n_3 c} A_1 A_2 e^{i\Delta k z}, \tag{2.2.10}
\]
Coupled Wave Equations (for SFG)

\[
\frac{dA_3}{dz} = \frac{2id_{\text{eff}}\omega_3}{n_3 c} A_1 A_2 e^{i\Delta k z},
\]

(2.2.10)

\[
\frac{dA_1}{dz} = \frac{2id_{\text{eff}}\omega_1}{n_1 c} A_3 A_2^* e^{-i\Delta k z},
\]

(2.2.12a)

\[
\frac{dA_2}{dz} = \frac{2id_{\text{eff}}\omega_2}{n_2 c} A_3 A_1^* e^{-i\Delta k z}.
\]

(2.2.12b)
Solving the Coupled Wave Equations (for SFG)

2.6 Sum-Frequency Generation

In Section 2.2, we treated the process of sum-frequency generation in the simple limit in which the two input fields are undepleted by the nonlinear interaction. In the present section, we treat this process more generally. We assume the configuration shown in Fig. 2.6.1.

\[d_{\text{eff}} = \frac{1}{2} \chi^{(2)}\]

\[\omega_1 \rightarrow \omega_1\]
\[\omega_2 \rightarrow \omega_2\]
\[\omega_3 \rightarrow \omega_3 = \omega_1 + \omega_2\]

FIGURE 2.6.1: Sum-frequency generation. Typically, no input field is applied at frequency \(\omega_3\).
Begin with a solution for the process where the following assumptions are made:

- \( \omega_1 \), \( \omega_2 \) are incident on a material, where \( \omega_1 \) is weak and \( \omega_2 \) is very intense
- Assume that the amplitude, \( A_2 \) is unaffected by the interaction

\[ d_{\text{eff}} = \frac{1}{2} \chi^{(2)} \]
Solving the Coupled Wave Equations (for SFG)

The coupled equations describing this case are:

\[
\frac{dA_1}{dz} = K_1 A_3 e^{-i \Delta k z}, \quad (2.6.1a)
\]

\[
\frac{dA_3}{dz} = K_3 A_1 e^{+i \Delta k z}, \quad (2.6.1b)
\]

where we have introduced the quantities

\[
K_1 = \frac{2i \omega_1^2 d_{\text{eff}}}{k_1 c^2} A_2^*, \quad K_3 = \frac{2i \omega_3^2 d_{\text{eff}}}{k_3 c^2} A_2, \quad (2.6.2a)
\]
Solving the Coupled Wave Equations (for SFG)

The solution to Eqs. (2.6.1) is particularly simple if we set $\Delta k = 0$, and we treat this case first. We take the derivative of Eq. (2.6.1a) to obtain

$$\frac{d^2 A_1}{dz^2} = K_1 \frac{dA_3}{dz}.$$  \hspace{1cm} (2.6.3)

We now use Eq. (2.6.1b) to eliminate $dA_3/dz$ from the right-hand side of this equation to obtain an equation involving only $A_1(z)$:

$$\frac{d^2 A_1}{dz^2} = -\kappa^2 A_1,$$  \hspace{1cm} (2.6.4)

Where: $\kappa^2 \equiv -K_1 K_3$
Solving the Coupled Wave Equations (for SFG)

\[
\frac{d^2 A_1}{dz^2} = -\kappa^2 A_1, \quad (2.6.4)
\]

The general solution to Eq. (2.6.4) is

\[
A_1(z) = B \cos \kappa z + C \sin \kappa z. \quad (2.6.6a)
\]

\[
A_3(z) = \frac{-B \kappa}{K_1} \sin \kappa z + \frac{C \kappa}{K_1} \cos \kappa z. \quad (2.6.6b)
\]
Solving the Coupled Wave Equations (for SFG)

\( A_1(0) \) specified. We find from Eq. (2.6.6b) that the boundary condition \( A_3(0) = 0 \) implies that \( C = 0 \), and from Eq. (2.6.6a) that \( B = A_1(0) \). The solution for the \( \omega_1 \) field is thus given by

\[
A_1(z) = A_1(0) \cos \kappa z.
\]

(2.6.7)

and for the \( \omega_3 \) field by

\[
A_3(z) = -A_1(0) \frac{\kappa}{K_1} \sin \kappa z.
\]

(2.6.8)

Which simplifies to the following result when we plug in the expressions for our defined constants \( K_1 \) and \( k \):

\[
A_3(z) = i \left( \frac{n_1 \omega_3}{n_3 \omega_1} \right)^{1/2} A_1(0) \sin \kappa z e^{i \phi_2}.
\]

(2.6.9)
Solving the Coupled Wave Equations (for SFG)

\[ A_1(z) = A_1(0) \cos \kappa z , \quad A_3(z) = i \left( \frac{n_1 \omega_3}{n_3 \omega_1} \right)^{1/2} A_1(0) \sin \kappa z e^{i \phi_2} \]

This solution is shown graphically in the following figure:

![Graph](image)

**FIGURE 2.6.2**: Variation of $|A_1|^2$ and $|A_3|^2$ for the case of perfect phase matching in the undepleted-pump approximation.