CHAPTER 6

REVIEW OF NONLINEAR OPTICAL PHENOMENA

In this chapter we deal with fundamental nonlinear optical phenomena, such as the generation of harmonics, parametric oscillation and amplification, frequency conversion, self-focusing, Raman, Brillouin and hyper-Raman scattering and multiphoton absorption and emission; we also provide a brief review of resonant coherent phenomena, such as superradiance, self-induced transparency and photon echoes. A more detailed quantum statistical treatment is then given in Chapter 10. The treatment of this chapter is rather brief and introductory and further details concerning the traditional description of nonlinear optical phenomena can be found in the monographs and reviews mentioned in the Introduction.

We consider nonlinear interactions of various orders of nonlinearity and we distinguish between those nonlinear optical interactions where the medium plays only a parametric role (optical parametric processes) and nonlinear interactions where the variables of the active medium occur explicitly (Raman scattering, multiphoton absorption and emission, etc.).

6.1 General classical description

The classical phenomenological description of nonlinear optical phenomena is based on the Maxwell equations for the electric and magnetic field vectors \( \mathbf{E} \) and \( \mathbf{H} \) in the nonlinear medium. By the standard method we can obtain from them the equivalent wave equation, usually for \( \mathbf{E} \), in the nonlinear medium

\[
\Delta \mathbf{E}(\mathbf{x}, t) - \mu_0 \varepsilon_0 \frac{\partial^2 \mathbf{E}(\mathbf{x}, t)}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}(\mathbf{x}, t)}{\partial t^2},
\]

\[
\nabla \cdot \mathbf{E}(\mathbf{x}, t) = - \frac{1}{\varepsilon_0} \nabla \cdot \mathbf{P}(\mathbf{x}, t),
\]

where \( \varepsilon_0 \) and \( \mu_0 \) are the permittivity and permeability constants of the vacuum; a non-magnetic and non-conducting medium is assumed, without external charges. If the intensity of the radiation is sufficiently high, the medium has a nonlinear response to the radiation, and the generalized electric polarization vector \( \mathbf{P} \) can be
written in the frequency domain as

\[
\mathbf{P}(\omega_i) = \chi^{(1)}(\omega_i) \cdot \mathbf{E}(\omega_i) + \sum_{j,k} \chi^{(2)}(\omega_i = \omega_j + \omega_k) \cdot \mathbf{E}(\omega_j) \mathbf{E}(\omega_k) + \\
+ \sum_{j,k,l} \chi^{(3)}(\omega_i = \omega_j + \omega_k + \omega_l) \cdot \mathbf{E}(\omega_j) \mathbf{E}(\omega_k) \mathbf{E}(\omega_l) + \ldots,
\]

(6.2)

where \(\chi^{(n)}\) are the susceptibility tensors of order \((n + 1)\), and multiple scalar product are expressed. Note that in the time domain, (6.2) is represented by a multifold convolution. Now we can treat the nonlinear processes of various orders, classified on the basis of (6.2). Since \(\mathbf{E}(t)\) and \(\mathbf{P}(t)\) are real vectors, \(\chi^{(n)}\) must fulfill the cross-symmetry conditions, such as \(\chi^{(1)}(\omega) = \chi^{(1)}(-\omega)\), etc.

### 6.2 The second-order phenomena

The second-order nonlinear optical phenomena are characterized by the second term in (6.2). Assuming propagation along the z-axis, and monochromatic waves, we obtain from (6.1a) for the parametric interaction of three waves, provided that \(k \mathbf{d} \mathbf{E}_j/\mathbf{d}z \gg d^2 \mathbf{E}_j/\mathbf{d}z^2\) (\(k = |\mathbf{k}|\),

\[
\begin{align*}
\frac{d\mathbf{E}_{1i}}{dz} &= \frac{i\omega_1}{2} \left( \frac{\mu_0}{\varepsilon_1} \right)^{1/2} \sum_{j,k} \chi^{(2)}_{ijk} \mathbf{E}_{3j} \mathbf{E}_{2k}^* \exp (i \Delta k z), \\
\frac{d\mathbf{E}_{2k}}{dz} &= \frac{i\omega_2}{2} \left( \frac{\mu_0}{\varepsilon_2} \right)^{1/2} \sum_{i,j} \chi^{(2)}_{kij} \mathbf{E}_{1i} \mathbf{E}_{3j}^* \exp (i \Delta k z), \\
\frac{d\mathbf{E}_{3j}}{dz} &= \frac{i\omega_3}{2} \left( \frac{\mu_0}{\varepsilon_3} \right)^{1/2} \sum_{i,k} \chi^{(2)}_{ijk} \mathbf{E}_{1i} \mathbf{E}_{2k} \exp (-i \Delta k z),
\end{align*}
\]

(6.3a) - (6.3c)

where \(i, j, k\) denote the cartesian components, \(\omega_j = \mathbf{k}_j/|\mu_0\varepsilon_j|^{1/2}\) (\(\varepsilon_j\) are permittivities) and \(\Delta k = k_3^{(i)} - k_2^{(i)} - k_1^{(i)}\) represents the phase mismatch, \(k_j^{(i)}\) being the \(i\)th polarization component of the wave vector of the \(j\)th wave. Further, the frequency resonance condition \(\omega_3 = \omega_1 + \omega_2\) holds.

If one cannot distinguish between waves 1 and 2, this three-wave interaction reduces to the degenerate case, described by the following set of coupled equations

\[
\begin{align*}
\frac{d\mathbf{E}_{1i}}{dz} &= i\omega_1 \left( \frac{\mu_0}{\varepsilon_1} \right)^{1/2} \sum_{j,k} \chi^{(2)}_{ikj} \mathbf{E}_{1j} \mathbf{E}_{2j}^* \exp (i \Delta k z), \\
\frac{d\mathbf{E}_{2j}}{dz} &= i\omega_2 \left( \frac{\mu_0}{\varepsilon_2} \right)^{1/2} \sum_{i,k} \chi^{(2)}_{jik} \mathbf{E}_{1i} \mathbf{E}_{2k} \exp (-i \Delta k z),
\end{align*}
\]

(6.4a) - (6.4b)

where \(\Delta k = k_3^{(i)} - 2k_1^{(i)}\) and \(\omega_2 = 2\omega_1\). Compared to (6.3), where the subfrequency modes are identified as 1 and 2 and the sum-frequency mode as 3, we have here denoted the subfrequency mode as 1 and the sum-frequency mode as 2.

Thus equations (6.3) describe, in various channels, the process of sum-frequency generation, if radiation of frequency \(\omega_3 = \omega_1 + \omega_2\) is generated from the subfre-