Introduction

Nonlinear optics is the study of the response of dielectric media to strong optical fields. The fields are sufficiently strong that the response of the medium is, as its name implies, nonlinear. That is, the polarization, which is the dipole moment per unit volume in the medium, is not a linear function of the applied electric field. In the equation for the polarization, there is a linear term, but, in addition, there are terms containing higher powers of the electric field. This leads to significant new types of behavior, one of the most notable being that frequencies different from that of the incident electromagnetic wave, such as harmonics or subharmonics, can be generated. Linear media do not change the frequency of light incident upon them. The first observation of a nonlinear optical effect was, in fact, second-harmonic generation – a laser beam entering a nonlinear medium produced a second beam at twice the frequency of the original. Another type of behavior that becomes possible in nonlinear media is that the index of refraction, rather than being a constant, is a function of the intensity of the light. For a light beam with a nonuniform intensity profile, this can lead to self-focusing of the beam.

Most nonlinear optical effects can be described using classical electromagnetic fields, and, in fact, the initial theory of nonlinear optics was formulated assuming the fields were classical. When the fields are quantized, however, a number of new effects emerge. Quantized fields are necessary if we want to describe fields that originate from spontaneous emission. For example, in a process known as spontaneous parametric down-conversion, a beam of light at one frequency, the pump, produces a beam at half the original frequency, the signal. This second beam is a result of spontaneous emission. The quantum properties of the down-converted beam are novel, a result of the fact that its photons are produced in pairs, one pump photon disappearing to produce, simultaneously, two signal photons. This leads to strong correlations between pairs of photons in the signal beam. In particular, the photons produced in this way can be quantum mechanically entangled. In addition, the signal beam can have smaller phase fluctuations than is possible with classical light. Both of these properties have made light produced by parametric down-conversion useful for applications in the field of quantum information.

The quantization of electrodynamics in nonlinear media is not straightforward, and so we will treat the canonical quantization of fields in some detail. Field quantization is a subject that is treated in just a few pages in most books on quantum optics, but here we will be much more thorough. We will discuss two approaches to this problem. The first is the quantization of the macroscopic Maxwell equations. The goal here is to obtain a quantized theory that has the macroscopic Maxwell equations as its Heisenberg equations of motion. The second approach is to make a model for the medium and quantize the entire matter–field system. Once this is done, an effective Hamiltonian describing the behavior of the fields in the medium can be found.

Many nonlinear optical systems can be discussed by employing only a few modes, and we shall employ this approach in discussing a number of simple systems. This will allow us to discuss some of the quantum mechanical correlations of the fields that these systems produce. Attributes such as squeezing and entanglement are properties that quantum fields can have but that classical fields cannot. As we shall see, quantum fields with these unusual features are produced by nonlinear optical systems. We initially explore the properties of these systems in free space, but we then move on to see what happens when they are placed in an optical cavity.

In order to discuss systems in cavities, we need to present the theory of open quantum systems and some of the mathematical techniques that have been developed to treat them. The nonlinear interaction couples a small number of cavity modes either to themselves or to each other, but these modes are coupled to modes outside the cavity through an output mirror. The external modes can be treated as a reservoir. Thus, our discussion of nonlinear devices in cavities will entail the introduction of reservoirs, operator Langevin and master equations, and techniques for turning these operator equations into c-number equations that can be more easily solved. It will also entail an input–output theory to relate the properties of the field inside the cavity to those of the field outside the cavity. It is the field outside the cavity, of course, that is usually measured.

It is also possible to treat more complicated systems, such as a field propagating in a nonlinear fiber. As well being the backbone of modern communications systems, optical fibers can generate strong nonlinear and quantum effects. They support the existence of quantum solitons, and their output fields can demonstrate squeezing and polarization squeezing. All of these effects have been demonstrated experimentally. In comparing theory to experiment, it is necessary to take into account the quantum noise sources in fibers, and we show how this can be done. This allows a detailed and quantitative test of the theoretical techniques explained here. It is also a demonstration of the quantum dynamics of a many-body system, since, as we shall see, fiber optics is equivalent to a system of interacting bosons in one dimension.

We conclude with a short chapter on the applications of nonlinear optics to the field of quantum information. We show how a degenerate parametric amplifier can be used to approximately clone quantum states, and how the squeezed states that are produced by such a device can be used to teleport them. We explain how these quantum states can be used to demonstrate the Einstein–Podolsky–Rosen paradox, and to generate a violation of the Bell inequality, issues that are important for fundamental physics. Understanding these issues and how they can be experimentally tested requires an understanding of both quantum mechanics and nonlinear optics.

The reader for whom this book is intended is a graduate student who has taken one-year graduate courses in electromagnetic theory and quantum theory. Essential results from these areas are summarized where needed. We do not assume any knowledge of quantum optics or quantum field theory. We also hope that physicists working in other fields will find the book useful.

We would like to emphasize that this textbook is not a review article, so we have made no attempt to provide a comprehensive bibliography of the field. We provide limited lists of additional reading at the ends of the chapters. This text is a result of our having worked in this field, and it emphasizes the points of view we have developed in doing so. There are certainly many other ways in which the quantum theory of nonlinear optics can be approached, and, in some cases, these are presented in textbooks by other authors.

Our presentation begins with a very brief survey of some topics in the classical theory on nonlinear optics. It is useful to see some of the basic ideas in a simpler classical context before jumping into the more complicated quantum case. The first chapter will provide a rather quick overview that will nonetheless serve as a foundation for what follows.

1 Classical nonlinear optics

Before discussing nonlinear optics with quantized fields, it is useful to have a look at what happens with classical electromagnetic fields in nonlinear dielectric media. The theory of nonlinear optics was originally developed using classical fields by Armstrong, Bloembergen, Ducuing and Pershan in 1962, stimulated by an experiment by Franken, Hill, Peters and Weinreich in which a second harmonic of a laser field was produced by shining the laser into a crystal. This classical theory is sufficient for many applications. For the most part, quantized fields were introduced later, although a quantum theory for the parametric amplifier, a nonlinear device in which three modes are coupled, was developed by Louisell, Yariv and Siegman as early as 1961. In any case, a study of the classical theory will give us an idea of some of the effects to look for when we formulate the more complicated quantum theory.

What we will present here is a very short introduction to the subject. Our intent is to use the classical theory to present some of the basic concepts and methods of nonlinear optics. Further information can be found in the list of additional reading at the end of the chapter. The discussion here is based primarily on the presentations in the books by N. Bloembergen and by R. W. Boyd.

1.1 Linear polarizability

We wish to survey some of the effects caused by the linear polarizability of a dielectric medium. When an electric field $\mathbf{E}(\mathbf{r}, t)$ is applied to a dielectric medium, a polarization, that is, a dipole moment per unit volume, is created in the medium. Maxwell's equations for a nonmagnetic material, but with the polarization included, are

$$\begin{aligned} \nabla \cdot \mathbf{D} &= 0, & \nabla \cdot \mathbf{B} &= 0, \\ \nabla \times \mathbf{E} &= -\frac{\partial \mathbf{B}}{\partial t}, & \nabla \times \mathbf{H} &= \frac{\partial \mathbf{D}}{\partial t}. \end{aligned} \tag{1.1}$$

Here $\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}$ is the displacement field, and $\mathbf{B} = \mu \mathbf{H}$ is the magnetic field. We use the bold notation $\mathbf{D} = (D_1, D_2, D_3)$ to indicate a 3-vector field at position $\mathbf{r} = (x, y, z)$ and time t, and generally omit the space-time arguments of fields for brevity. We use SI units, so ϵ_0 is the vacuum permittivity, and μ is the magnetic permeability. Here we separate the polarizability term so that we can more readily analyze nonlinear effects.

Differentiating the equation for $\nabla \times \mathbf{H}$ with respect to time and making use of the equation for $\nabla \times \mathbf{E}$ gives

$$\nabla \times \nabla \times \mathbf{E} = -\mu \frac{\partial^2 \mathbf{D}}{\partial t^2}. \tag{1.2}$$

An alternative form, in terms of the polarization field, is

$$\nabla \times \nabla \times \mathbf{E} + \frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = -\mu \frac{\partial^2 \mathbf{P}}{\partial t^2}. \tag{1.3}$$

Here $c = 1/\sqrt{\mu\epsilon_0}$. Examining Eq. (1.3), we first note that $\nabla \times \nabla \times \mathbf{E} = \nabla(\nabla \cdot \mathbf{E}) - \nabla^2\mathbf{E}$. In free space, $\nabla \cdot \mathbf{E} = 0$, but this is no longer true in a medium. In many cases it is, however, small and can be neglected. For example, if the field is close to a plane wave, this term will be small. We shall assume that it can be neglected in most situations we consider, which leads to the form of the wave equation most commonly used in nonlinear optics:

$$\nabla^2 \mathbf{E} - \frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu \frac{\partial^2 \mathbf{P}}{\partial t^2}. \tag{1.4}$$

In typical cases of interest, $\mu \approx \mu_0$, where μ_0 is the vacuum permeability, so that c is the vacuum light velocity to a good approximation. We retain the full permeability in the Maxwell equations for generality, as there are small contributions to the magnetic permeability – typically of $O(10^{-6})$ – in dielectric materials.

1.1.1 Linear polarizability

If the field is not too strong, the response of the medium is linear. This means that the polarization, $\mathbf{P}$, is linear in the applied field, and, in general,

$$P_j = \epsilon_0[\boldsymbol{\chi}^{(1)} \cdot \mathbf{E}]_j = \epsilon_0 \sum_{k=1}^{3} \chi^{(1)}_{jk} E_k. \tag{1.5}$$

In this equation, $[\boldsymbol{\chi}^{(1)}]_{ij} = \chi^{(1)}_{ij}$ is the linear susceptibility tensor of the medium. This tells us that the polarization acts as a source for the field, and, in particular, if the polarization has terms oscillating at a particular frequency, then those terms will give rise to components of the field oscillating at the same frequency. For a linear dielectric medium with no significant magnetization, the permeability equals the vacuum permeability. In a linear, isotropic medium, we can omit the tensor subscripts, writing $\mathbf{P} = \epsilon_0\chi^{(1)}\mathbf{E}$. Since

$$\mathbf{D} = \boldsymbol{\epsilon} \cdot \mathbf{E} = \epsilon_0\mathbf{E} + \mathbf{P}, \tag{1.6}$$

it follows that the electric permittivity in a dielectric is given by

$$\boldsymbol{\epsilon} = \epsilon_0[1 + \boldsymbol{\chi}^{(1)}], \tag{1.7}$$

and the exact linear wave equation, Eq. (1.2), reduces to

$$\nabla \times \nabla \times (\boldsymbol{\epsilon}^{-1} \cdot \mathbf{D}) = -\mu \frac{\partial^2 \mathbf{D}}{\partial t^2}. \tag{1.8}$$

An alternative form, valid for homogeneous, isotropic dielectrics, is obtained from Eq. (1.4), which simplifies to

$$\nabla^2 \mathbf{E} = \frac{1}{v_p^2} \frac{\partial^2 \mathbf{E}}{\partial t^2}, \tag{1.9}$$

where $v_p = 1/\sqrt{\mu\epsilon}$ is the phase velocity of electromagnetic waves in the medium.

The results in this chapter will generally assume an input of a plane-wave, nearly monochromatic, laser beam with polarization $\hat{\mathbf{e}}$, angular frequency $\omega = 2\pi f$, wavevector $k = 2\pi/\lambda$, and slowly varying envelope $\mathcal{E}$ in time,

$$\mathbf{E}(\mathbf{r}, t) = \mathcal{E}(\mathbf{r}, t)\hat{\mathbf{e}}\, e^{-i\omega t} + c.c. \tag{1.10}$$

(where $c.c.$ denotes complex conjugate). For an envelope that is slowly varying in time *and* space, we introduce

$$\mathbf{E}(\mathbf{r}, t) = \mathcal{A}(\mathbf{r}, t)\hat{\mathbf{e}}\, e^{-i(\omega t - kx)} + c.c. \tag{1.11}$$

If $\mathbf{P} = 0$ and $\mu = \mu_0$, the resulting speed of light in the vacuum is $c = 1/\sqrt{\epsilon_0 \mu_0}$. More generally, the phase velocity of electromagnetic radiation in the dielectric medium is v_p, and is given by

$$v_p = \frac{1}{\sqrt{\mu\epsilon}} = \frac{c}{n_r} = \frac{\omega}{k}. \tag{1.12}$$

Here n_r is the refractive index, which is given in terms of the linear susceptibility by

$$n_r = \sqrt{1 + \chi^{(1)}}. \tag{1.13}$$

Inserting Eq. (1.11) into Eq. (1.9), and dropping second-derivative terms in time and the propagation, that is, the x, direction (that these terms are small follows from the assumption that the envelope varies slowly compared to the wavelength and optical frequency), we find that

$$\left[\frac{\partial}{\partial x} + \frac{1}{v_p}\frac{\partial}{\partial t} - \frac{i}{2k}\nabla_\perp^2\right]\mathcal{A}(\mathbf{r}, t) = 0. \tag{1.14}$$

Here, $\nabla_\perp^2 \equiv \partial^2/\partial y^2 + \partial^2/\partial z^2$. This equation is called the paraxial wave equation. It has characteristic traveling plane-wave solutions of the form $f(x - v_p t)$, consisting of waveforms traveling at the phase velocity, v_p. When a transverse variation is included, this equation leads to focusing and diffraction effects. Here $\lambda f = v_p = c/n_r$, in terms of the frequency f and wavelength λ. Since $\nabla \times \mathbf{E} = -\partial \mathbf{B}/\partial t$, it follows that the corresponding magnetic field is

$$\mathbf{B}(\mathbf{r}, t) = (\mathcal{A}/v_p)\hat{\mathbf{k}} \times \hat{\mathbf{e}}\, e^{-i(\omega t - kx)} + c.c. \tag{1.15}$$

Let us note that our definition of complex amplitudes follows the convention of Glauber. In some texts, complex amplitudes are defined as $\mathbf{E}(\mathbf{r}, t) = \Re[\mathcal{E}_C(\mathbf{r}, t)\hat{\mathbf{e}}\, e^{i\omega t}]$. These are related to ours by $\mathcal{E}_C = 2\mathcal{E}$, and give rise to differences of powers of 2^{n-1} in the nonlinear equations for an nth-order nonlinearity in later sections.

1.1.2 Energy density, intensity and power

The dispersionless energy density $\mathcal{H}$ has the usual classical form of

$$\mathcal{H} = \frac{1}{2}[\epsilon|\mathbf{E}|^2(t) + \mu|\mathbf{H}|^2(t)]. \tag{1.16}$$

Defining $\mathcal{H}_{av}$ as the time-averaged energy density, the magnetic and electric field contributions to the energy of a plane-wave solution to Maxwell's equations are equal, and the total intensity for a dispersionless medium is

$$I_0 = v_p \mathcal{H}_{av} = 2v_p\epsilon|\mathcal{A}(\mathbf{r}, t)|^2. \tag{1.17}$$

More rigorously, we demonstrate later that there are dispersive corrections to the above results for energy, intensity and power, due to the frequency dependence of the dielectric response. These are assumed negligible for simplicity here.

Lasers have a transverse envelope function $u(\mathbf{r})$ that is typically Gaussian, with a beam radius or 'waist' of W_0 that varies in the x direction. At a beam focus, this depends primarily on the transverse coordinate $\mathbf{r}_\perp = (y, z)$, so that

$$\mathcal{E}(\mathbf{r}_\perp) = \mathcal{E}(0)e^{-|\mathbf{r}_\perp|^2/W_0^2}. \tag{1.18}$$

Integrating over the beam waist, the total laser power is

$$P = \frac{1}{2}\pi I_0 W_0^2 = \pi v_p \epsilon W_0^2 |\mathcal{E}|^2. \tag{1.19}$$

We will generally ignore transverse effects. However, these are important in understanding how beam intensities, which cause nonlinear effects, are related to laser powers. We will treat the more general case of dispersive energy later in this chapter.

1.2 Nonlinear polarizability

If the field is sufficiently strong, the linear relation breaks down and nonlinear terms must be taken into account. In Bloembergen's approach, we expand the polarization in a Taylor expansion in $\mathbf{E}$ to give

$$P_j = \epsilon_0\left[\sum_k \chi_{jk}^{(1)} E_k + \sum_{k,l} \chi_{jkl}^{(2)} E_k E_l + \sum_{k,l,m} \chi_{jklm}^{(3)} E_k E_l E_m + \cdots\right]. \tag{1.20}$$

Here, we have kept the first three terms in the power series expansion of the polarization in terms of the field. The quantities $\chi^{(2)}$ and $\chi^{(3)}$ are the second- and third-order nonlinear susceptibilities, respectively. We should also note that Eq. (1.20) is often written as a matrix or generalized tensor multiplication, in the form

$$\begin{aligned}\mathbf{P}(\mathbf{E}) &= \mathbf{P}^L + \mathbf{P}^{NL} \\ &= \epsilon_0 \sum_{n>0} \boldsymbol{\chi}^{(n)} : \mathbf{E}^{\otimes n},\end{aligned} \tag{1.21}$$

where $\mathbf{P}^L \equiv \epsilon_0 \boldsymbol{\chi}^{(1)} \cdot \mathbf{E} = (\boldsymbol{\epsilon} - 1) \cdot \mathbf{E}$ is the linear response, while $\mathbf{P}^{NL}$ is the nonlinear polarization. We use the notation $\mathbf{E}^{\otimes n}$ to indicate a vector Kronecker product, mapping a vector into an nth-order tensor, so

$$[\mathbf{E}^{\otimes n}]_{i_1 \ldots i_n} \equiv E_{i_1} \cdots E_{i_n}. \tag{1.22}$$

We can use the $\boldsymbol{\chi}$ coefficients to expand the displacement field $\mathbf{D}$ directly in terms of $\mathbf{E}$, which simplifies results in later chapters. We define $\boldsymbol{\epsilon}^{(1)} = \boldsymbol{\epsilon}$ and $\boldsymbol{\epsilon}^{(n)} = \epsilon_0 \boldsymbol{\chi}^{(n)}$, so that $\boldsymbol{\epsilon}^{(n)}$ becomes an $(n+1)$th-order tensor. Then

$$\mathbf{D}(\mathbf{E}) = \sum_{n>0} \boldsymbol{\epsilon}^{(n)} : \mathbf{E}^{\otimes n}. \tag{1.23}$$

Not all the terms in this series are necessarily present. The even terms like $\chi^{(2)}$ are only present if the medium is not invariant under spatial inversion ($\mathbf{r} \to -\mathbf{r}$). This follows from the fact that, if the medium is invariant under spatial inversion, the $\chi^{(2)}$ for the inverted medium will be the same as that for the original medium, i.e. under spatial inversion, we will have $\chi^{(2)} \to \chi^{(2)}$. However, under spatial inversion, we also have that $\mathbf{P} \to -\mathbf{P}$ and $\mathbf{E} \to -\mathbf{E}$. Consequently, while $\mathbf{P} \to -\mathbf{P}$ implies that we should have

$$\chi^{(2)} : \mathbf{E} \otimes \mathbf{E} \to -\chi^{(2)} : \mathbf{E} \otimes \mathbf{E}, \tag{1.24}$$

the relations $\chi^{(2)} \to \chi^{(2)}$ and $\mathbf{E} \to -\mathbf{E}$ show us that instead we have

$$\chi^{(2)} : \mathbf{E} \otimes \mathbf{E} \to \chi^{(2)} : \mathbf{E} \otimes \mathbf{E}. \tag{1.25}$$

The only way these conditions can be consistent is if $\chi^{(2)} = 0$. Therefore, for many materials, we do, in fact, have $\chi^{(2)} = 0$, and all even terms vanish for the same reason. Then, the first nonzero nonlinear susceptibility is $\chi^{(3)}$. The other symmetry properties of the medium also directly affect the susceptibilities. For example, in the common case of an amorphous solid with complete spherical symmetry, the susceptibilities are diagonal matrices and tensors. In this case, we refer to them as scalars, and one can simply drop the indices. Similarly, for plane-polarized radiation, under conditions where the polarization is always in the same plane, it is also possible to ignore the indices, though for a different reason.

We can also define an inverse permittivity tensor, $\boldsymbol{\eta}^{(n)}$, as a coefficient of a power series expansion of the macroscopic *electric* field in terms of the macroscopic displacement field. This greatly simplifies the treatment of quantized fields. In the subsequent chapters on quantization, we will make use of this expansion, which has the form

$$\mathbf{E}(\mathbf{D}) = \sum_{n>0} \boldsymbol{\eta}^{(n)} : \mathbf{D}^{\otimes n}. \tag{1.26}$$

This is an equally valid approach to nonlinear response, as these are simply two alternative power series expansions.

It is possible to express the inverse permittivity tensors $\eta^{(j)}$ in terms of the permittivities $\epsilon^{(j)}$. Let us assume that we know the $\chi^{(n)}$, and hence the $\epsilon^{(n)}$ coefficients already. Combining the two expansions, we have

$$\mathbf{E} = \sum_{n>0} \boldsymbol{\eta}^{(n)} : \left[\sum_{m>0} \boldsymbol{\epsilon}^{(m)} : \mathbf{E}^{\otimes m} \right]^{\otimes n}. \tag{1.27}$$

We now simply equate equal powers of **E**, so that formally:

$$\begin{aligned} \mathbf{1} &= \boldsymbol{\eta}^{(1)} : \boldsymbol{\epsilon}^{(1)}, \\ \mathbf{0} &= \boldsymbol{\eta}^{(2)} : \boldsymbol{\epsilon}^{(1)}\boldsymbol{\epsilon}^{(1)} + \boldsymbol{\eta}^{(1)} : \boldsymbol{\epsilon}^{(2)}, \\ \mathbf{0} &= \boldsymbol{\eta}^{(3)} : \boldsymbol{\epsilon}^{(1)}\boldsymbol{\epsilon}^{(1)}\boldsymbol{\epsilon}^{(1)} + \boldsymbol{\eta}^{(2)} : \boldsymbol{\epsilon}^{(2)}\boldsymbol{\epsilon}^{(1)} + \boldsymbol{\eta}^{(2)} : \boldsymbol{\epsilon}^{(1)}\boldsymbol{\epsilon}^{(2)} + \boldsymbol{\eta}^{(1)} : \boldsymbol{\epsilon}^{(3)}. \end{aligned} \tag{1.28}$$

Writing this out in detail (and recalling that we define $\eta_{ij} = \eta_{ij}^{(1)}$), we see that, for the lowest-order terms,

$$\eta_{ij} = [\epsilon^{-1}]_{ij}, \qquad \eta_{jnp}^{(2)} = -\eta_{jk}\epsilon_{klm}^{(2)}\eta_{ln}\eta_{mp}, \tag{1.29}$$

where we introduce the Einstein summation convention, in which repeated indices are summed over, and we note that there will be contributions from all the terms $\epsilon^{(1)}, \ldots, \epsilon^{(n)}$ to the inverse nonlinear coefficient $\eta^{(n)}$.

1.2.1 Second-order nonlinearity

We can calculate the effects of nonlinearities by substituting the response functions, i.e. the expansions for the polarization of the medium, into Maxwell's equations. In our initial survey of nonlinear optical effects, we shall ignore all indices, and treat all quantities as scalars; this corresponds to an assumption of plane polarization in a single direction. Let us first look at second-order nonlinearities. If the applied field oscillates at frequency ω,

$$E(t) = \mathcal{E}_0[e^{i\omega t} + e^{-i\omega t}] = 2\mathcal{E}_0 \cos \omega t, \tag{1.30}$$

then the nonlinear part of the polarization, P^{NL}, will be

$$P^{NL}(t) = \epsilon_0 \chi^{(2)} E(t)^2 = 2\epsilon_0 \chi^{(2)} \mathcal{E}_0^2 (1 + \cos 2\omega t). \tag{1.31}$$

The polarization has a term oscillating at twice the applied frequency, and this will give rise to a field whose frequency is also 2ω. This process is known as second-harmonic generation. It can be, and is, used to double the frequency of the output of a laser by sending the beam through an appropriate material, that is, one with a nonzero value of $\chi^{(2)}$. As was mentioned earlier, this was the first nonlinear optical effect that was observed.

Now suppose our applied field oscillates at two frequencies:

$$E(t) = 2[\mathcal{E}_1 \cos \omega_1 t + \mathcal{E}_2 \cos \omega_2 t]. \tag{1.32}$$

The nonlinear polarization is then

$$\begin{aligned} P^{NL}(t) = 2\epsilon_0 \chi^{(2)} \{ & \mathcal{E}_1^2 (1 + \cos 2\omega_1 t) + \mathcal{E}_2^2 (1 + \cos 2\omega_2 t) \\ & + 2\mathcal{E}_1 \mathcal{E}_2 [\cos(\omega_1 + \omega_2)t + \cos(\omega_1 - \omega_2)t] \}. \end{aligned} \tag{1.33}$$

In this case, not only do we have terms oscillating at twice the frequencies of the components of the applied field, but we also have terms oscillating at the sum and difference of their frequencies. These processes are called sum- and difference-frequency generation, respectively.

1.2.2 Third-order nonlinearity

Let us move on to a third-order nonlinearity. For an applied field oscillating at a single frequency, as before, we find that (assuming that $\chi^{(2)} = 0$)

$$P^{NL}(t) = 2\epsilon_0 \chi^{(3)} \mathcal{E}_0^3 [\cos 3\omega t + 3 \cos \omega t]. \tag{1.34}$$

The first term will clearly cause a field at 3ω, the third harmonic of the applied field, to be generated. In order to see the effect of the second term, it is useful to combine the linear and nonlinear parts of the polarization to get the total polarization,

$$P(t) = 2\epsilon_0 (\chi^{(1)} + 3\chi^{(3)} \mathcal{E}_0^2) \mathcal{E}_0 \cos \omega t + 2\epsilon_0 \chi^{(3)} \mathcal{E}_0^3 \cos 3\omega t. \tag{1.35}$$

When there is no nonlinear polarization, the polarization is proportional to the field, and the constant of proportionality, $\chi^{(1)}$, is directly related to the refractive index of the material. When there is a nonlinearity, we see that the component of the polarization at the same frequency as the applied field is similar to what it is in the linear case, except that

$$\chi^{(1)} \to \chi^{(1)} + 3\chi^{(3)} \mathcal{E}_0^2. \tag{1.36}$$

This results in a refractive index that depends on the intensity of the applied field according to Eqs (1.13) and (1.17). The refractive index can therefore be written as

$$n_r(I) = \sqrt{1 + \chi^{(1)} + 3\chi^{(3)} I/(2v_p \epsilon)}. \tag{1.37}$$

This can be expanded as a power series in the intensity, so that, to lowest order,

$$n_r(I) = n_1 + n_2 I + \cdots , \tag{1.38}$$

where the nonlinear refractive index, n_2, is given by

$$n_2 = \frac{3\chi^{(3)}}{4\epsilon c}. \tag{1.39}$$

This is often called the Kerr effect, after its original discoverer.

1.3 Frequency dependence and dispersion

So far we have assumed that the response of the medium to an applied field, that is, the polarization at time t, depends only on the electric field at time t. This is, of course, an