Nonlinear Optics

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ACADEMIC PRESS, INC.
Harcourt Brace Jovanovich, Publishers
Boston San Diego New York
London Sydney Tokyo Toronto

This book is printed on acid-free paper. ∞

ACADEMIC PRESS, INC.
1250 Sixth Avenue, San Diego, CA 92101

United Kingdom Edition published by
ACADEMIC PRESS LIMITED
24-28 Oval Road, London NW1 7DX

Library of Congress Cataloging-in-Publication Data:

Boyd, Robert W., date.
Nonlinear optics / Robert W. Boyd.
p. cm.
Includes bibliographical references and index.
ISBN 0-12-121680-2 (alk. paper)
1. Nonlinear optics. I. Title.
QC446.2.B69 1992 91-9903
535.2—dc20 CIP

PRINTED IN THE UNITED STATES OF AMERICA

91 92 93 94 9 8 7 6 5 4 3 2 1

Preface

Nonlinear optics is the study of the interaction of intense laser light with matter. This book is a textbook on nonlinear optics at the level of a beginning graduate student. The intent of the book is to provide an introduction to the field of nonlinear optics that stresses fundamental concepts and that enables the student to go on to perform independent research in this field. The author has successfully used a preliminary version of this book in his course at the University of Rochester, which is typically attended by students ranging from seniors to advanced PhD students from disciplines that include optics, physics, chemistry, electrical engineering, mechanical engineering, and chemical engineering. This book could be used in graduate courses in the areas of nonlinear optics, quantum optics, quantum electronics, laser physics, electrooptics, and modern optics. By deleting some of the more difficult sections, this book would also be suitable for use by advanced undergraduates. On the other hand, some of the material in the book is rather advanced and would be suitable for senior graduate students and research scientists.

The field of nonlinear optics is now thirty years old, if we take its beginnings to be the observation of second-harmonic generation by Franken and coworkers in 1961. Interest in this field has grown continuously since its beginnings, and the field of nonlinear optics now ranges from fundamental studies of the interaction of light with matter to applications such as laser frequency conversion and optical switching. In fact, the field of nonlinear optics has grown so enormously that it is not possible for one book to cover all of the topics of current interest. In addition, since I want this book to be accessible to beginning graduate students, I have attempted to treat the topics that are covered in a reasonably self-contained manner. This consideration also restricts the number of topics that can be treated. My strategy in deciding what topics to include has been to stress the fundamental aspects of nonlinear optics, and to include applications and experimental results only as necessary to illustrate these fundamental issues. Many of the specific topics that I have chosen to include are those of particular historical value.

Nonlinear optics is notationally very complicated, and unfortunately much of the notational complication is unavoidable. Because the notational aspects of nonlinear optics have historically been very confusing, considerable effort is made, especially in the early chapters, to explain the notational conventions. The book uses primarily the Gaussian system of units, both to establish a connection with the historical papers of nonlinear optics, most of which were written using the Gaussian system, and also because the author believes that the laws of electromagnetism are more physically transparent when written in this system. At several places in the text (see especially the appendices at the end of the book), tables are provided to facilitate conversion to other systems of units.

The book is organized as follows: Chapter 1 presents an introduction to the field of nonlinear optics from the perspective of the nonlinear susceptibility. The nonlinear susceptibility is a quantity that is used to determine the nonlinear polarization of a material medium in terms of the strength of an applied optical-frequency electric field. It thus provides a framework for describing nonlinear optical phenomena. Chapter 2 continues the description of nonlinear optics by describing the propagation of light waves through nonlinear optical media by means of the optical wave equation. This chapter introduces the important concept of phase matching and presents detailed descriptions of the important nonlinear optical phenomena of second-harmonic generation and sum- and difference-frequency generation. Chapter 3 concludes the introductory portion of the book by presenting a description of the quantum mechanical theory of the nonlinear optical susceptibility. Simplified expressions for the nonlinear susceptibility are first derived through use of the Schrödinger equation, and then more accurate expressions are derived through use of the density matrix equations of motion. The density matrix formalism is itself developed in considerable detail in this chapter in order to render this important discussion accessible to the beginning student.

Chapters 4 through 6 deal with properties and applications of the nonlinear refractive index. Chapter 4 introduces the topic of the nonlinear refractive index. Properties, including tensor properties, of the nonlinear refractive index are discussed in detail, and physical processes that lead to the nonlinear refractive index, such as nonresonant electronic polarization and molecular orientation, are described. Chapter 5 is devoted to a description of nonlinearities in the refractive index resulting from the response of two-level atoms. Related topics that are discussed in this chapter include saturation, power broadening, optical Stark shifts, Rabi oscillations, and dressed atomic states. Chapter 6 deals with applications of the nonlinear refractive index. Topics that are included are optical phase conjugation, self focusing, optical bistability, two-beam coupling, pulse propagation, and the formation of optical solitons.

Chapters 7 through 9 deal with spontaneous and stimulated light scattering and the related topic of acoustooptics. Chapter 7 introduces this area by presenting a description of theories of spontaneous light scattering and by describing the important practical topic of acousto-optics. Chapter 8 presents a description of stimulated Brillouin and stimulated Rayleigh scattering. These topics are related in that they both entail the scattering of light from material disturbances that can be described in terms of the standard thermodynamic variables of pressure and entropy. Also included in this chapter is a description of phase conjugation by stimulated Brillouin scattering and a theoretical description of stimulated Brillouin scattering in gases. Chapter 9 presents a description of stimulated Raman and stimulated Rayleigh-wing scattering. These processes are related in that they entail the scattering of light from disturbances associated with the positions of atoms within a molecule.

The book concludes with Chapter 10, which treats the electrooptic and photorefractive effects. The chapter begins with a description of the electrooptic effect and describes how this effect can be used to fabricate light modulators. The chapter then presents a description of the photorefractive effect, which is a nonlinear optical interaction that results from the electrooptic effect. The use of the photorefractive effect in two-beam coupling and in four-wave mixing is also described.

The author wishes to acknowledge his deep appreciation for discussions of the material in this book with his graduate students at the University of Rochester. He is sure that he has learned as much from them as they have from him. He also gratefully acknowledges discussions with numerous other professional colleagues, including N. Bloembergen, D. Chemla, R. Y. Chiao, J. H. Eberly, C. Flytzanis, J. Goldhar, G. Grynberg, J. H. Haus, R. W. Hellwarth, K. R. MacDonald, S. Mukamel, P. Narum, M. G. Raymer, J. E. Sipe, C. R. Stroud, Jr., C. H. Townes, H. Winful, and B. Ya. Zel'dovich. In addition, the assistance of J. J. Maki and A. Gamliel in the preparation of the figures is gratefully acknowledged.

Contents

Chapter 1

The Nonlinear Optical Susceptibility

1.1. Introduction to Nonlinear Optics

Nonlinear optics is the study of phenomena that occur as a consequence of the modification of the optical properties of a material system by the presence of light. Typically, only laser light is sufficiently intense to modify the optical properties of a material system. In fact, the beginning of the field of nonlinear optics is often taken to be the discovery of second-harmonic generation by Franken *et al.* in 1961, shortly after the demonstration of the first working laser by Maiman in 1960. Nonlinear optical phenomena are "nonlinear" in the sense that they occur when the response of a material system to an applied optical field depends in a nonlinear manner upon the strength of the optical field. For example, second-harmonic generation occurs as a result of the part of the atomic response that depends quadratically on the strength of the applied optical field. Consequently, the intensity of the light generated at the second-harmonic frequency tends to increase as the square of the intensity of the applied laser light.

In order to describe more precisely what we mean by an optical nonlinearity, let us consider how the dipole moment per unit volume, or polarization $\tilde{P}(t)$, of a material system depends upon the strength $\tilde{E}(t)$ of the applied optical field.* In the case of conventional (i.e., linear) optics, the induced

* Throughout the text, we use the tilde to denote a quantity that varies rapidly in time. Constant quantities, slowly varying quantities, and Fourier amplitudes are written without the tilde. See, for example, Eq. (1.2.1).

polarization depends linearly upon the electric field strength in a manner that can often be described by the relationship

$$\tilde{P}(t) = \chi^{(1)}\tilde{E}(t), \tag{1.1.1}$$

where the constant of proportionality $\chi^{(1)}$ is known as the linear susceptibility. In nonlinear optics, the nonlinear optical response can often be described by generalizing Eq. (1.1.1) by expressing the polarization $\tilde{P}(t)$ as a power series in the field strength $\tilde{E}(t)$ as

$$\begin{aligned} \tilde{P}(t) &= \chi^{(1)}\tilde{E}(t) + \chi^{(2)}\tilde{E}^2(t) + \chi^{(3)}\tilde{E}^3(t) + \cdots \\ &\equiv \tilde{P}^{(1)}(t) + \tilde{P}^{(2)}(t) + \tilde{P}^{(3)}(t) + \cdots. \end{aligned} \tag{1.1.2}$$

The quantities $\chi^{(2)}$ and $\chi^{(3)}$ are known as the second- and third-order nonlinear optical susceptibilities, respectively. For simplicity, we have taken the fields $\tilde{P}(t)$ and $\tilde{E}(t)$ to be scalar quantities in writing Eqs. (1.1.1) and (1.1.2). In Section 1.3 we show how to treat the vector nature of the fields; in such a case $\chi^{(1)}$ becomes a second-rank tensor, $\chi^{(2)}$ becomes a third-rank tensor, etc. In writing Eqs. (1.1.1) and (1.1.2) in the form shown, we have also assumed that the polarization at time t depends only on the instantaneous value of the electric field strength. The assumption that the medium responds instantaneously also implies (through the Kramers–Kronig relations)* that the medium must be lossless and dispersionless. We shall see in Section 1.3 how to generalize these equations for the case of a medium with dispersion and loss. In general, the nonlinear susceptibilities depend on the frequencies of the applied fields, but under our present assumption of instantaneous response we take them to be constants.

We shall refer to $\tilde{P}^{(2)}(t) = \chi^{(2)}\tilde{E}(t)^2$ as the second-order nonlinear polarization and to $\tilde{P}^{(3)}(t) = \chi^{(3)}\tilde{E}(t)^3$ as the third-order nonlinear polarization. We shall see later in this section that the physical processes that occur as a result of the second-order polarization $\tilde{P}^{(2)}$ are distinct from those that occur as a result of the third-order polarization $\tilde{P}^{(3)}$. In addition, we shall show in Section 1.5 that second-order nonlinear optical interactions can occur only in noncentrosymmetric crystals, that is, in crystals that do not display inversion symmetry. Since liquids, gases, amorphous solids (such as glass), and even many crystals do display inversion symmetry, $\chi^{(2)}$ vanishes identically for such media, and consequently they cannot produce second-order nonlinear optical interactions. On the other hand, third-order nonlinear optical interac-

* See, for example, Loudon (1983) for a discussion of the Kramers–Kronig relations.

tions (i.e., those described by a $\chi^{(3)}$ susceptibility) can occur both for centrosymmetric and noncentrosymmetric media.

We shall see in later sections of this book how to calculate the values of the nonlinear susceptibilities for various physical mechanisms that can lead to optical nonlinearities. For the present, we shall make a simple order-of-magnitude estimate of the size of these quantities for the common case in which the nonlinearity is electronic in origin. One might expect that the lowest-order correction term $\tilde{P}^{(2)}$ would be comparable to the linear response $\tilde{P}^{(1)}$ when the amplitude of the applied field strength $\tilde{E}$ was of the order of the characteristic atomic electric field strength $E_{at} = e/a_0^2$, where $-e$ is the charge of the electron and $a_0 = \hbar^2/me^2$ is the Bohr radius of the hydrogen atom (here $\hbar$ is Planck's constant divided by 2π, and m is the mass of the electron). Numerically, we find that $E_{at} = 2 \times 10^7$ esu.* We thus expect that under conditions of nonresonant excitation the second-order susceptibility $\chi^{(2)}$ will be of the order of $\chi^{(1)}/E_{at}$. For condensed matter $\chi^{(1)}$ is of the order of unity, and we hence expect that $\chi^{(2)}$ will be of the order of $1/E_{at}$, or that

$$\chi^{(2)} \simeq 5 \times 10^{-8} \text{ esu} = 5 \times 10^{-8} \frac{\text{cm}}{\text{statvolt}}. \tag{1.1.3}$$

Similarly, we expect $\chi^{(3)}$ to be of the order of $\chi^{(1)}/E_{at}^2$, which for condensed matter is of the order of

$$\chi^{(3)} \simeq 3 \times 10^{-15} \text{ esu} = 3 \times 10^{-15} \frac{\text{cm}^2}{\text{statvolt}^2}. \tag{1.1.4}$$

The most common procedure for describing nonlinear optical phenomena is based on expressing the polarization $\tilde{P}(t)$ in terms of the applied electric field strength $\tilde{E}(t)$, as we have done in Eq. (1.1.2). The reason why the polarization plays a key role in the description of nonlinear optical phenomena is that a time-varying polarization can act as the source of new components of the electromagnetic field. For example, we shall see in Section 2.1 that the wave equation in nonlinear optical media often has the form

$$\nabla^2\tilde{E} - \frac{n^2}{c^2}\frac{\partial^2\tilde{E}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2\tilde{P}}{\partial t^2}, \tag{1.1.5}$$

* Except where otherwise noted, we use the gaussian system of units in this book. Following standard conventions, we usually do not give the dimensions of physical quantities explicitly, but instead quote values simply in gaussian units or (equivalently for all cases treated herein) in electrostatic units, abbreviated esu. For the present case, the dimensions of E_{at} are statvolt/cm. See also the discussion in the appendix to this book on the conversion between systems of units.

where n is the refractive index and c is the speed of light in vacuum. We can interpret this expression as an inhomogeneous wave equation in which the polarization $\tilde{P}$ drives the electric field $\tilde{E}$. This equation expresses the fact that, whenever $\partial^2\tilde{P}/\partial t^2$ is nonzero, charges are being accelerated, and according to Larmor's theorem from electromagnetism, accelerated charges generate electromagnetic radiation.

1.2. Descriptions of Nonlinear Optical Interactions

In the present section, we present brief qualitative descriptions of a number of nonlinear optical interactions. In addition, for those processes that can occur in a lossless medium, we indicate how they can be described in terms of the nonlinear contributions to the polarization described by Eq. (1.1.2).* Our motivation is to provide the reader with an indication of the variety of nonlinear optical phenomena that can occur. These interactions are described in greater detail in later sections of this book. In this section we also introduce some notational conventions and some of the basic concepts of nonlinear optics.

Second-Harmonic Generation

As an example of a nonlinear optical interaction, let us consider the process of second-harmonic generation, which is illustrated schematically in Fig. 1.2.1.

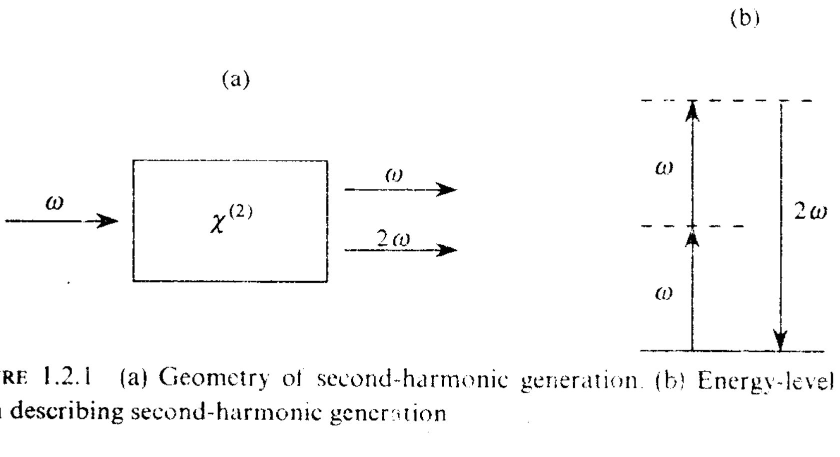

FIGURE 1.2.1 (a) Geometry of second-harmonic generation. (b) Energy-level diagram describing second-harmonic generation

* Recall that Eq. (1.1.2) is valid only for a medium that is lossless and dispersionless.

Here a laser beam whose electric field strength is represented as

$$\tilde{E}(t) = Ee^{-i\omega t} + \text{c.c.} \tag{1.2.1}$$

is incident upon a crystal for which the second-order susceptibility $\chi^{(2)}$ is nonzero. The nonlinear polarization that is created in such a crystal is given according to Eq. (1.1.2) as $\tilde{P}^{(2)}(t) = \chi^{(2)}\tilde{E}^2(t)$ or as

$$\tilde{P}^{(2)}(t) = 2\chi^{(2)}EE^* + (\chi^{(2)}E^2e^{-2i\omega t} + \text{c.c.}). \tag{1.2.2}$$

We see that the second-order polarization consists of a contribution at zero frequency (the first term) and a contribution at frequency 2ω (the second term). According to the driven wave equation (1.1.5), this latter contribution can lead to the generation of radiation at the second-harmonic frequency. Note that the first contribution in Eq. (1.2.2) does not lead to the generation of electromagnetic radiation (because its second time derivative vanishes); it leads to a process known as optical rectification in which a static electric field is created within the nonlinear crystal.

Under proper experimental conditions, the process of second-harmonic generation can be so efficient that nearly all of the power in the incident radiation at frequency ω is converted to radiation at the second-harmonic frequency 2ω. One common use of second-harmonic generation is to convert the output of a fixed-frequency laser into a different spectral region. For example, the Nd:YAG laser operates in the near infrared at a wavelength of 1.06 μm. Second-harmonic generation is routinely used to convert the wavelength of the radiation to 0.53 μm, in the middle of the visible spectrum.

Second-harmonic generation can also be visualized by considering the interaction in terms of the exchange of photons between the various frequency components of the field. According to this picture, which is illustrated in part (b) of Fig. 1.2.1, two photons of frequency ω are destroyed and a photon of frequency 2ω is simultaneously created in a single quantum-mechanical process. The solid line in the figure represents the atomic ground state, and the dashed lines represent what are known as virtual levels. These levels are not energy eigenlevels of the free atom, but rather represent the combined energy of one of the energy eigenstates of the atom and of one or more photons of the radiation field.

The theory of second-harmonic generation is developed more fully in Section 2.6.

Sum- and Difference-Frequency Generation

Let us next consider the circumstance in which the optical field incident upon a nonlinear optical medium characterized by a nonlinear susceptibility $\chi^{(2)}$ consists of two distinct frequency components, which we represent in the form

$$\tilde{E}(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + \text{c.c.} \tag{1.2.3}$$

Then, assuming as in Eq. (1.1.2) that the second-order contribution to the nonlinear polarization is of the form

$$\tilde{P}^{(2)}(t) = \chi^{(2)}\tilde{E}(t)^2, \tag{1.2.4}$$

we find that the nonlinear polarization is given by

$$\begin{aligned}\tilde{P}^{(2)}(t) = \chi^{(2)}[E_1^2 e^{-2i\omega_1 t} + E_2^2 e^{-i2\omega_2 t} + 2E_1E_2 e^{-(\omega_1+\omega_2)t} \\ + 2E_1E_2^* e^{-i(\omega_1-\omega_2)t} + \text{c.c.}] + 2\chi^{(2)}[E_1E_1^* + E_2E_2^*].\end{aligned} \tag{1.2.5}$$

It is convenient to express this result using the notation

$$\tilde{P}^{(2)}(t) = \sum_n P(\omega_n)e^{-i\omega_n t}, \tag{1.2.6}$$

where the summation extends over positive and negative frequencies ω_n. The complex amplitudes of the various frequency components of the nonlinear polarization are hence given by

$$\begin{aligned}P(2\omega_1) &= \chi^{(2)}E_1^2 \quad \text{(SHG)},\\ P(2\omega_2) &= \chi^{(2)}E_2^2 \quad \text{(SHG)}\\ P(\omega_1+\omega_2) &= 2\chi^{(2)}E_1E_2 \quad \text{(SFG)},\\ P(\omega_1-\omega_2) &= 2\chi^{(2)}E_1E_2^* \quad \text{(DFG)},\\ P(0) &= 2\chi^{(2)}(E_1E_1^* + E_2E_2^*) \quad \text{(OR)}.\end{aligned} \tag{1.2.7}$$

Here we have labeled each expression by the name of the physical process that it describes, such as second-harmonic generation (SHG), sum-frequency generation (SFG), difference-frequency generation (DFG), and optical rectification (OR). Note that, in accordance with our complex notation, there is also a response at the negative of each of the nonzero frequencies given above:

$$\begin{aligned}P(-2\omega_1) &= \chi^{(2)}E_1^{*2}, \qquad & P(-2\omega_2) &= \chi^{(2)}E_2^{*2},\\ P(-\omega_1-\omega_2) &= 2\chi^{(2)}E_1^*E_2^* \qquad & P(\omega_2-\omega_1) &= 2\chi^{(2)}E_2E_1^*.\end{aligned} \tag{1.2.8}$$

However, since each of these quantities is simply the complex conjugate of one of the quantities given in Eq. (1.2.7), it is not necessary to take explicit account of both the positive and negative frequency components.*

We see from Eq. (1.2.7) that four different nonzero frequency components are present in the nonlinear polarization. However, typically no more than one of these frequency components will be present with any appreciable intensity in the radiation generated by the nonlinear optical interaction. The reason for this behavior is that the nonlinear polarization can efficiently produce an output signal only if a certain phase-matching condition (which is discussed in detail in Section 2.7) is satisfied, and usually this condition cannot be satisfied for more than one frequency component of the nonlinear polarization. Operationally, one often chooses which frequency component will be radiated by properly selecting the polarization of the input radiation and orientation of the nonlinear crystal.

Sum-Frequency Generation

Let us now consider the process of sum-frequency generation, which is illustrated in Fig. 1.2.2. According to Eq. (1.2.7), the complex amplitude of the nonlinear polarization describing this process is given by the expression

$$P(\omega_1 + \omega_2) = 2\chi^{(2)}E_1E_2. \tag{1.2.9}$$

In many ways the process of sum-frequency generation is analogous to that of second-harmonic generation, except that in sum-frequency generation the two input waves are at different frequencies. One application of sum-frequency generation is to produce tunable radiation in the ultraviolet spectral region by

* Not all workers in nonlinear optics use our convention that the fields and polarizations are given by Eqs. (1.2.3) and (1.2.6). Another common convention is to define the field amplitudes according to

$$\tilde{E}(t) = \tfrac{1}{2}(E'_1e^{-i\omega_1t} + E'_2e^{-i\omega_2t} + \text{c.c.}),$$

$$\tilde{P}(t) = \tfrac{1}{2}\sum_n P'(\omega_n)e^{-i\omega_nt},$$

where in the second expression the summation extends over all positive and negative frequencies. Using this convention, one finds that

$$P'(2\omega_1) = \tfrac{1}{2}\chi^{(2)}E_1'^2, \qquad P'(2\omega_2) = \tfrac{1}{2}\chi^{(2)}E_2'^2,$$

$$P'(\omega_1 + \omega_2) = \chi^{(2)}E'_1E'_2, \qquad P'(\omega_1 - \omega_2) = \chi^{(2)}E'_1E_2'^*,$$

$$P'(0) = \chi^{(2)}(E'_1E_1'^* + E'_2E_2'^*).$$

Note that these expressions differ from Eqs. (1.2.7) by factors of $\frac{1}{2}$.