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Nonlinear Optics

THIRD EDITION

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Third Edition

Robert W. Boyd

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Preface to the Third Edition

It has been a great pleasure for me to have prepared the latest edition of my book on nonlinear optics. My intrigue in the subject matter of this book is as strong as it was when the first edition was published in 1992.

The principal changes present in the third edition are as follows: (1) The book has been entirely rewritten using the SI system of units. I personally prefer the elegance of the gaussian system of units, which was used in the first two editions, but I realize that most readers would prefer the SI system, and the change was made for this reason. (2) In addition, a large number of minor changes have been made throughout the text to clarify the intended meaning and to make the arguments easier to follow. I am indebted to the countless comments received from students and colleagues both in Rochester and from around the world that have allowed me to improve the writing in this manner. (3) Moreover, several sections that treat entirely new material have been added. Applications of harmonic generation, including applications within the fields of microscopy and biophotonics, are treated in Subsection 2.7.1. Electromagnetically induced transparency is treated in Section 3.8. Some brief but crucial comments regarding limitations to the maximum size of the intensity-induced refractive-index change are made in Section 4.7. The use of nonlinear optical methods for inducing unusual values of the group velocity of light are discussed briefly in Section 3.8 and in Subsection 6.6.2. Spectroscopy based on coherent anti–Stokes Raman scattering (CARS) is discussed in Section 10.5. In addition, the appendix has been expanded to include brief descriptions of both the SI and gaussian systems of units and procedures for conversion between them.

The book in its present form contains far too much material to be covered within a conventional one-semester course. For this reason, I am often asked for advice on how to structure a course based on the content of my textbook. Some of my thoughts along these lines are as follows: (1) I have endeavored as much as possible to make each part of the book self-contained. Thus, the sophisticated reader can read the book in any desired order and can read only sections of personal interest. (2) Nonetheless, when using the book as a course text, I suggest starting with Chapters 1 and 2, which present the basic formalism of the subject material. At that point, topics of interest can be taught in nearly any order. (3) Special mention should be made regarding Chapters 3 and 6, which deal with quantum mechanical treatments of nonlinear optical phenomena. These chapters are among the most challenging of any within the book. These chapters can be skipped entirely if one is comfortable with establishing only a phenomenological description of nonlinear optical phenomena. Alternatively, these chapters can form the basis of a formal treatment of how the laws of quantum mechanics can be applied to provide detailed descriptions of a variety of optical phenomena. (4) From a different perspective, I am sometimes asked for my advice on extracting the essential material from the book—that is, in determining which are topics that everyone should know. This question often arises in the context of determining what material students should study when preparing for qualifying exams. My best response to questions of this sort is that the essential material is as follows: Chapter 1 in its entirety; Sections 2.1–2.3, 2.4, and 2.10 of Chapter 2; Subsection 3.5.1 of Chapter 3; Sections 4.1, 4.6, and 4.7 of Chapter 4; Chapter 7 in its entirety; Section 8.1 of Chapter 8; and Section 9.1 of Chapter 9. (5) Finally, I often tell my classroom students that my course is in some ways as much a course on optical physics as it is a course on nonlinear optics. I simply use the concept of nonlinear optics as a unifying theme for presenting conceptual issues and practical applications of optical physics. Recognizing that this is part of my perspective in writing, this book could be useful to its readers.

I want to express my thanks once again to the many students and colleagues who have given me useful advice and comments regarding this book over the past fifteen years. I am especially indebted to my own graduate students for the assistance and encouragement they have given to me.

Robert Boyd
Rochester, New York
October, 2007

Preface to the Second Edition

In the ten years since the publication of the first edition of this book, the field of nonlinear optics has continued to achieve new advances both in fundamental physics and in practical applications. Moreover, the author's fascination with this subject has held firm over this time interval. The present work extends the treatment of the first edition by including a considerable body of additional material and by making numerous small improvements in the presentation of the material included in the first edition.

The primary differences between the first and second editions are as follows.

Two additional sections have been added to Chapter 1, which deals with the nonlinear optical susceptibility. Section 1.6 deals with time-domain descriptions of optical nonlinearities, and Section 1.7 deals with Kramers–Kronig relations in nonlinear optics. In addition, a description of the symmetry properties of gallium arsenide has been added to Section 1.5.

Three sections have been added to Chapter 2, which treats wave-equation descriptions of nonlinear optical interactions. Section 2.8 treats optical parametric oscillators, Section 2.9 treats quasi-phase-matching, and Section 2.11 treats nonlinear optical surface interactions.

Two sections have been added to Chapter 4, which deals with the intensity-dependent refractive index. Section 4.5 treats thermal nonlinearities, and Section 4.6 treats semiconductor nonlinearities.

Chapter 5 is an entirely new chapter dealing with the molecular origin of the nonlinear optical response. (Consequently the chapter numbers of all the following chapters are one greater than those of the first edition.) This chapter treats electronic nonlinearities in the static approximation, semiempirical models of the nonlinear susceptibility, the nonlinear response of conjugated polymers, the bond charge model of optical nonlinearities, nonlinear optics of chiral materials, and nonlinear optics of liquid crystals.

In Chapter 7 on processes resulting from the intensity-dependent refractive index, the section on self-action effects (now Section 7.1) has been significantly expanded. In addition, a description of optical switching has been included in Section 7.3, now entitled optical bistability and optical switching.

In Chapter 9, which deals with stimulated Brillouin scattering, a discussion of transient effects has been included.

Chapter 12 is an entirely new chapter dealing with optical damage and multiphoton absorption. Chapter 13 is an entirely new chapter dealing with ultrafast and intense-field nonlinear optics.

The Appendices have been expanded to include a treatment of the gaussian system of units. In addition, many additional homework problems and literature references have been added.

I would like to take this opportunity to thank my many colleagues who have given me advice and suggestions regarding the writing of this book. In addition to the individuals mentioned in the preface to the first edition, I would like to thank G. S. Agarwal, P. Agostini, G. P. Agrawal, M. D. Feit, A. L. Gaeta, D. J. Gauthier, L. V. Hau, F. Kajzar, M. Kauranen, S. G. Lukishova, A. C. Melissinos, Q-H. Park, M. Saffman, B. W. Shore, D. D. Smith, I. A. Walmsley, G. W. Wicks, and Z. Zyss. I especially wish to thank M. Kauranen and A. L. Gaeta for suggesting additional homework problems and to thank A. L. Gaeta for advice on the preparation of Section 13.2.

Preface to the First Edition

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 acoustooptics. 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. The beginning of the field of nonlinear optics is often taken to be the discovery of second-harmonic generation by Franken *et al.* (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 on the strength of the optical field. For example, second-harmonic generation occurs as a result of the part of the atomic response that scales quadratically with 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 on the strength $\tilde{E}(t)$ of an applied optical

* It should be noted, however, that some nonlinear effects were discovered prior to the advent of the laser. The earliest example known to the authors is the observation of saturation effects in the luminescence of dye molecules reported by G.N. Lewis *et al.* (1941).

field.* In the case of conventional (i.e., linear) optics, the induced polarization depends linearly on the electric field strength in a manner that can often be described by the relationship

$$\tilde{P}(t) = \epsilon_0 \chi^{(1)} \tilde{E}(t), \tag{1.1.1}$$

where the constant of proportionality $\chi^{(1)}$ is known as the linear susceptibility and ϵ_0 is the permittivity of free space. In nonlinear optics, the 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

$$\begin{aligned} \tilde{P}(t) &= \epsilon_0\left[\chi^{(1)}\tilde{E}(t) + \chi^{(2)}\tilde{E}^2(t) + \chi^{(3)}\tilde{E}^3(t) + \cdots\right] \\ &\equiv \tilde{P}^{(1)}(t) + \tilde{P}^{(2)}(t) + \tilde{P}^{(3)}(t) + \cdots. \end{aligned} \tag{1.1.2}$$

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, and so on. In writing Eqs. (1.1.1) and (1.1.2) in the forms 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 $\tilde{P}^{(2)}(t) = \epsilon_0 \chi^{(2)} \tilde{E}^2(t)$ as the second-order nonlinear polarization and to $\tilde{P}^{(3)}(t) = \epsilon_0 \chi^{(3)} \tilde{E}^3(t)$ as the third-order nonlinear polarization. We shall see later in this section that physical processes that occur as a result of the second-order polarization $\tilde{P}^{(2)}$ tend to be 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),

* 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).

† See, for example, Loudon (1973, Chapter 4) or the discussion in Section 1.7 of this book for a discussion of the Kramers–Kronig relations.