

Nonlinear Optics

Third Edition

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Contents

<i>Preface to the Third Edition</i>	xiii
<i>Preface to the Second Edition</i>	xv
<i>Preface to the First Edition</i>	xvii
1. The Nonlinear Optical Susceptibility	1
1.1. Introduction to Nonlinear Optics	1
1.2. Descriptions of Nonlinear Optical Processes	4
1.3. Formal Definition of the Nonlinear Susceptibility	17
1.4. Nonlinear Susceptibility of a Classical Anharmonic Oscillator	21
1.5. Properties of the Nonlinear Susceptibility	33
1.6. Time-Domain Description of Optical Nonlinearities	52
1.7. Kramers–Kronig Relations in Linear and Nonlinear Optics	58
Problems	63
References	65
2. Wave-Equation Description of Nonlinear Optical Interactions	69
2.1. The Wave Equation for Nonlinear Optical Media	69
2.2. The Coupled-Wave Equations for Sum-Frequency Generation	74
2.3. Phase Matching	79
2.4. Quasi-Phase-Matching	84
2.5. The Manley–Rowe Relations	88
2.6. Sum-Frequency Generation	91
2.7. Second-Harmonic Generation	96
	vii

2.8.	Difference-Frequency Generation and Parametric Amplification	105
2.9.	Optical Parametric Oscillators	108
2.10.	Nonlinear Optical Interactions with Focused Gaussian Beams	116
2.11.	Nonlinear Optics at an Interface	122
	Problems	128
	References	132
3.	Quantum-Mechanical Theory of the Nonlinear Optical Susceptibility	135
3.1.	Introduction	135
3.2.	Schrödinger Calculation of Nonlinear Optical Susceptibility	137
3.3.	Density Matrix Formulation of Quantum Mechanics	150
3.4.	Perturbation Solution of the Density Matrix Equation of Motion	158
3.5.	Density Matrix Calculation of the Linear Susceptibility	161
3.6.	Density Matrix Calculation of the Second-Order Susceptibility	170
3.7.	Density Matrix Calculation of the Third-Order Susceptibility	180
3.8.	Electromagnetically Induced Transparency	185
3.9.	Local-Field Corrections to the Nonlinear Optical Susceptibility	194
	Problems	201
	References	204
4.	The Intensity-Dependent Refractive Index	207
4.1.	Descriptions of the Intensity-Dependent Refractive Index	207
4.2.	Tensor Nature of the Third-Order Susceptibility	211
4.3.	Nonresonant Electronic Nonlinearities	221
4.4.	Nonlinearities Due to Molecular Orientation	228
4.5.	Thermal Nonlinear Optical Effects	235
4.6.	Semiconductor Nonlinearities	240
4.7.	Concluding Remarks	247
	References	251
5.	Molecular Origin of the Nonlinear Optical Response	253
5.1.	Nonlinear Susceptibilities Calculated Using Time-Independent Perturbation Theory	253

5.2.	Semiempirical Models of the Nonlinear Optical Susceptibility	259
	Model of Boling, Glass, and Owyong	260
5.3.	Nonlinear Optical Properties of Conjugated Polymers	262
5.4.	Bond-Charge Model of Nonlinear Optical Properties	264
5.5.	Nonlinear Optics of Chiral Media	268
5.6.	Nonlinear Optics of Liquid Crystals	271
	Problems	273
	References	274
6.	Nonlinear Optics in the Two-Level Approximation	277
6.1.	Introduction	277
6.2.	Density Matrix Equations of Motion for a Two-Level Atom	278
6.3.	Steady-State Response of a Two-Level Atom to a Monochromatic Field	285
6.4.	Optical Bloch Equations	293
6.5.	Rabi Oscillations and Dressed Atomic States	301
6.6.	Optical Wave Mixing in Two-Level Systems	313
	Problems	326
	References	327
7.	Processes Resulting from the Intensity-Dependent Refractive Index	329
7.1.	Self-Focusing of Light and Other Self-Action Effects	329
7.2.	Optical Phase Conjugation	342
7.3.	Optical Bistability and Optical Switching	359
7.4.	Two-Beam Coupling	369
7.5.	Pulse Propagation and Temporal Solitons	375
	Problems	383
	References	388
8.	Spontaneous Light Scattering and Acousto-optics	391
8.1.	Features of Spontaneous Light Scattering	391
8.2.	Microscopic Theory of Light Scattering	396
8.3.	Thermodynamic Theory of Scalar Light Scattering	402

8.4.	Acoustooptics	413
	Problems	427
	References	428
9.	Stimulated Brillouin and Stimulated Rayleigh Scattering	429
9.1.	Stimulated Scattering Processes	429
9.2.	Electrostriction	431
9.3.	Stimulated Brillouin Scattering (Induced by Electrostriction)	436
9.4.	Phase Conjugation by Stimulated Brillouin Scattering	448
9.5.	Stimulated Brillouin Scattering in Gases	453
9.6.	Stimulated Brillouin and Stimulated Rayleigh Scattering	455
	Problems	468
	References	470
10.	Stimulated Raman Scattering and Stimulated Rayleigh-Wing Scattering	473
10.1.	The Spontaneous Raman Effect	473
10.2.	Spontaneous versus Stimulated Raman Scattering	474
10.3.	Stimulated Raman Scattering Described by the Nonlinear Polarization	479
10.4.	Stokes–Anti-Stokes Coupling in Stimulated Raman Scattering	488
10.5.	Coherent Anti-Stokes Raman Scattering	499
10.6.	Stimulated Rayleigh-Wing Scattering	501
	Problems	508
	References	508
11.	The Electrooptic and Photorefractive Effects	511
11.1.	Introduction to the Electrooptic Effect	511
11.2.	Linear Electrooptic Effect	512
11.3.	Electrooptic Modulators	516
11.4.	Introduction to the Photorefractive Effect	523
11.5.	Photorefractive Equations of Kukhtarev <i>et al.</i>	526
11.6.	Two-Beam Coupling in Photorefractive Materials	528
11.7.	Four-Wave Mixing in Photorefractive Materials	536
	Problems	540
	References	540

12. Optically Induced Damage and Multiphoton Absorption	543
12.1. Introduction to Optical Damage	543
12.2. Avalanche-Breakdown Model	544
12.3. Influence of Laser Pulse Duration	546
12.4. Direct Photoionization	548
12.5. Multiphoton Absorption and Multiphoton Ionization	549
Problems	559
References	559
13. Ultrafast and Intense-Field Nonlinear Optics	561
13.1. Introduction	561
13.2. Ultrashort Pulse Propagation Equation	561
13.3. Interpretation of the Ultrashort-Pulse Propagation Equation	567
13.4. Intense-Field Nonlinear Optics	571
13.5. Motion of a Free Electron in a Laser Field	572
13.6. High-Harmonic Generation	575
13.7. Nonlinear Optics of Plasmas and Relativistic Nonlinear Optics	579
13.8. Nonlinear Quantum Electrodynamics	583
Problem	586
References	586
Appendices	589
A. The SI System of Units	589
Further reading	596
B. The Gaussian System of Units	596
Further reading	600
C. Systems of Units in Nonlinear Optics	600
D. Relationship between Intensity and Field Strength	602
E. Physical Constants	603
Index	605

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.