Nonlinear Optics

Fourth Edition

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ACADEMIC PRESS
An imprint of Elsevier

Contents

Preface to the Fourth Edition ..... *xv*
Preface to the Third Edition ..... *xvii*
Preface to the Second Edition ..... *xix*
Preface to the First Edition ..... *xxi*

Chapter 1: The Nonlinear Optical Susceptibility *1*
1.1 Introduction to Nonlinear Optics 1
1.2 Descriptions of Nonlinear Optical Processes 4
1.2.1 Second-Harmonic Generation 4
1.2.2 Sum- and Difference-Frequency Generation 6
1.2.3 Sum-Frequency Generation 7
1.2.4 Difference-Frequency Generation 8
1.2.5 Optical Parametric Oscillation 9
1.2.6 Third-Order Nonlinear Optical Processes 10
1.2.7 Third-Harmonic Generation 10
1.2.8 Intensity-Dependent Refractive Index 11
1.2.9 Third-Order Interactions (General Case) 11
1.2.10 Parametric versus Nonparametric Processes 13
1.2.11 Saturable Absorption 14
1.2.12 Two-Photon Absorption 15
1.2.13 Stimulated Raman Scattering 16
1.3 Formal Definition of the Nonlinear Susceptibility 16
1.4 Nonlinear Susceptibility of a Classical Anharmonic Oscillator 20
1.4.1 Noncentrosymmetric Media 21
1.4.2 Miller's Rule 26
1.4.3 Centrosymmetric Media 27

1.5 Properties of the Nonlinear Susceptibility 32
1.5.1 Reality of the Fields 33
1.5.2 Intrinsic Permutation Symmetry 34
1.5.3 Symmetries for Lossless Media 34
1.5.4 Field Energy Density for a Nonlinear Medium 35
1.5.5 Kleinman's Symmetry 37
1.5.6 Contracted Notation 38
1.5.7 Effective Value of d (d_{eff}) 40
1.5.8 Spatial Symmetry of the Nonlinear Medium 41
1.5.9 Influence of Spatial Symmetry on the Linear Optical Properties of a Material Medium 41
1.5.10 Influence of Inversion Symmetry on the Second-Order Nonlinear Response 42
1.5.11 Influence of Spatial Symmetry on the Second-Order Susceptibility ... 44
1.5.12 Number of Independent Elements of $\chi^{(2)}_{ijk}(\omega_3, \omega_2, \omega_1)$ 45
1.5.13 Distinction between Noncentrosymmetric and Cubic Crystal Classes . 45
1.5.14 Distinction between Noncentrosymmetric and Polar Crystal Classes .. 50
1.5.15 Influence of Spatial Symmetry on the Third-Order Nonlinear Response 50
1.6 Time-Domain Description of Optical Nonlinearities 50
1.7 Kramers–Kronig Relations in Linear and Nonlinear Optics 56
1.7.1 Kramers–Kronig Relations in Linear Optics 56
1.7.2 Kramers–Kronig Relations in Nonlinear Optics 59
Problems 61
References 63

Chapter 2: Wave-Equation Description of Nonlinear Optical Interactions 65

2.1 The Wave Equation for Nonlinear Optical Media 65
2.2 The Coupled-Wave Equations for Sum-Frequency Generation 70
2.2.1 Phase-Matching Considerations 72
2.3 Phase Matching 74
2.4 Quasi-Phase-Matching (QPM) 79
2.5 The Manley–Rowe Relations 83
2.6 Sum-Frequency Generation 86
2.7 Second-Harmonic Generation 91
2.7.1 Applications of Second-Harmonic Generation 98
2.8 Difference-Frequency Generation and Parametric Amplification 100
2.9 Optical Parametric Oscillators 102
2.9.1 Influence of Cavity Mode Structure on OPO Tuning 105
2.10 Nonlinear Optical Interactions with Focused Gaussian Beams 109
2.10.1 Paraxial Wave Equation 109
2.10.2 Gaussian Beams 110

2.10.3 Harmonic Generation Using Focused Gaussian Beams 112
2.11 Nonlinear Optics at an Interface 116
2.12 Advanced Phase Matching Methods 121
Problems 130
References 134

Chapter 3: Quantum-Mechanical Theory of the Nonlinear Optical Susceptibility .. 137
3.1 Introduction 137
3.2 Schrödinger Equation Calculation of the Nonlinear Optical Susceptibility . 138
3.2.1 Energy Eigenstates 139
3.2.2 Perturbation Solution to Schrödinger's Equation 140
3.2.3 Linear Susceptibility 142
3.2.4 Second-Order Susceptibility 144
3.2.5 Third-Order Susceptibility 146
3.2.6 Third-Harmonic Generation in Alkali Metal Vapors 148
3.3 Density Matrix Formulation of Quantum Mechanics 151
3.3.1 Example: Two-Level Atom 158
3.4 Perturbation Solution of the Density Matrix Equation of Motion 159
3.5 Density Matrix Calculation of the Linear Susceptibility 161
3.5.1 Linear Response Theory 164
3.6 Density Matrix Calculation of the Second-Order Susceptibility 169
3.6.1 $\chi^{(2)}$ in the Limit of Nonresonant Excitation 178
3.7 Density Matrix Calculation of the Third-Order Susceptibility 179
3.8 Electromagnetically Induced Transparency 184
3.9 Local-Field Effects in the Nonlinear Optics 192
3.9.1 Local-Field Effects in Linear Optics 192
3.9.2 Local-Field Effects in Nonlinear Optics 194
Problems 198
References 201

Chapter 4: The Intensity-Dependent Refractive Index 203
4.1 Descriptions of the Intensity-Dependent Refractive Index 203
4.2 Tensor Nature of the Third-Order Susceptibility 209
4.2.1 Propagation through Isotropic Nonlinear Media 213
4.3 Nonresonant Electronic Nonlinearities 217
4.3.1 Classical, Anharmonic Oscillator Model of Electronic Nonlinearities . 218
4.3.2 Quantum-Mechanical Model of Nonresonant Electronic Nonlinearities 218
4.3.3 $\chi^{(3)}$ in the Low-Frequency Limit 222
4.4 Nonlinearities Due to Molecular Orientation 223
4.4.1 Tensor Properties of $\chi^{(3)}$ for the Molecular Orientation Effect 229

4.5 Thermal Nonlinear Optical Effects 231
4.5.1 Thermal Nonlinearities with Continuous-Wave Laser Beams 233
4.5.2 Thermal Nonlinearities with Pulsed Laser Beams 234
4.6 Semiconductor Nonlinearities 235
4.6.1 Nonlinearities Resulting from Band-to-Band Transitions 235
4.6.2 Nonlinearities Involving Virtual Transitions 241
4.7 Concluding Remarks 243
Problems 245
References 247

Chapter 5: Molecular Origin of the Nonlinear Optical Response 249

5.1 Nonlinear Susceptibilities Calculated Using Time-Independent Perturbation Theory 249
5.1.1 Hydrogen Atom 250
5.1.2 General Expression for the Nonlinear Susceptibility in the Quasi-Static Limit 251
5.2 Semiempirical Models of the Nonlinear Optical Susceptibility 255
Model of Boling, Glass, and Owyoung 256
5.3 Nonlinear Optical Properties of Conjugated Polymers 257
5.4 Bond-Charge Model of Nonlinear Optical Properties 259
5.5 Nonlinear Optics of Chiral Media 264
5.6 Nonlinear Optics of Liquid Crystals 266
Problems 269
References 269

Chapter 6: Nonlinear Optics in the Two-Level Approximation 273

6.1 Introduction 273
6.2 Density Matrix Equations of Motion for a Two-Level Atom 274
6.2.1 Closed Two-Level Atom 276
6.2.2 Open Two-Level Atom 279
6.2.3 Two-Level Atom with a Non-Radiatively Coupled Third Level 279
6.3 Steady-State Response of a Two-Level Atom to a Monochromatic Field ... 280
6.4 Optical Bloch Equations 288
6.4.1 Harmonic Oscillator Form of the Density Matrix Equations 291
6.4.2 Adiabatic-Following Limit 293
6.5 Rabi Oscillations and Dressed Atomic States 295
6.5.1 Rabi Solution of the Schrödinger Equation 296
6.5.2 Solution for an Atom Initially in the Ground State 298
6.5.3 Dressed States 302
6.5.4 Inclusion of Relaxation Phenomena 305

6.6 Optical Wave Mixing in Two-Level Systems 307
6.6.1 Solution of the Density Matrix Equations for a Two-Level Atom in the Presence of Pump and Probe Fields 308
6.6.2 Nonlinear Susceptibility and Coupled-Amplitude Equations 315
Problems 319
References 320

Chapter 7: Processes Resulting from the Intensity-Dependent Refractive Index 321

7.1 Self-Focusing of Light and Other Self-Action Effects 321
7.1.1 Self-Trapping of Light 324
7.1.2 Mathematical Description of Self-Action Effects 327
7.1.3 Laser Beam Breakup into Many Filaments 328
7.1.4 Self-Action Effects with Pulsed Laser Beams 333
7.2 Optical Phase Conjugation 334
7.2.1 Aberration Correction by Phase Conjugation 336
7.2.2 Phase Conjugation by Degenerate Four-Wave Mixing 338
7.2.3 Polarization Properties of Phase Conjugation 345
7.3 Optical Bistability and Optical Switching 349
7.3.1 Absorptive Bistability 351
7.3.2 Refractive Bistability 354
7.3.3 Optical Switching 356
7.4 Two-Beam Coupling 359
7.5 Pulse Propagation and Temporal Solitons 365
7.5.1 Self-Phase Modulation 365
7.5.2 Pulse Propagation Equation 368
7.5.3 Temporal Optical Solitons 372
Problems 374
References 379

Chapter 8: Spontaneous Light Scattering and Acoustooptics 381

8.1 Features of Spontaneous Light Scattering 381
8.1.1 Fluctuations as the Origin of Light Scattering 382
8.1.2 Scattering Coefficient 384
8.1.3 Scattering Cross Section 385
8.2 Microscopic Theory of Light Scattering 386
8.3 Thermodynamic Theory of Scalar Light Scattering 392
8.3.1 Ideal Gas 394
8.3.2 Spectrum of the Scattered Light 395
8.3.3 Brillouin Scattering 395
8.3.4 Stokes Scattering (First Term in Eq. (8.3.36)) 398

8.3.5 Anti-Stokes Scattering (Second Term in Eq. (8.3.36)) 400
8.3.6 Rayleigh Center Scattering 402
8.4 Acoustooptics 403
8.4.1 Bragg Scattering of Light by Sound Waves 403
8.4.2 Raman–Nath Effect 412
Problems 416
References 417

Chapter 9: Stimulated Brillouin and Stimulated Rayleigh Scattering 419
9.1 Stimulated Scattering Processes 419
9.2 Electrostriction 421
9.3 Stimulated Brillouin Scattering (Induced by Electrostriction) 425
9.3.1 Pump Depletion Effects in SBS 431
9.3.2 SBS Generator 433
9.3.3 Transient and Dynamical Features of SBS 436
9.4 Phase Conjugation by Stimulated Brillouin Scattering 437
9.5 Stimulated Brillouin Scattering in Gases 441
9.6 General Theory of Stimulated Brillouin and Stimulated Rayleigh Scattering 443
9.6.1 Appendix: Definition of the Viscosity Coefficients 454
Problems 456
References 457

Chapter 10: Stimulated Raman Scattering and Stimulated Rayleigh-Wing Scattering 459
10.1 The Spontaneous Raman Effect 459
10.2 Spontaneous versus Stimulated Raman Scattering 460
10.3 Stimulated Raman Scattering Described by the Nonlinear Polarization 465
10.4 Stokes–Anti-Stokes Coupling in Stimulated Raman Scattering 474
10.4.1 Dispersionless, Nonlinear Medium without Gain or Loss 478
10.4.2 Medium without a Nonlinearity 479
10.4.3 Stokes–Anti-Stokes Coupling in Stimulated Raman Scattering 480
10.5 Coherent Anti-Stokes Raman Scattering 483
10.6 Stimulated Rayleigh-Wing Scattering 486
10.6.1 Polarization Properties of Stimulated Rayleigh-Wing Scattering 490
Problems 492
References 492

Chapter 11: The Electrooptic and Photorefractive Effects 495
11.1 Introduction to the Electrooptic Effect 495
11.2 Linear Electrooptic Effect 496
11.3 Electrooptic Modulators 500

11.4 Introduction to the Photorefractive Effect 507
11.5 Photorefractive Equations of Kukhtarev et al. 508
11.6 Two-Beam Coupling in Photorefractive Materials 511
11.7 Four-Wave Mixing in Photorefractive Materials 518
11.7.1 Externally Self-Pumped Phase-Conjugate Mirror 519
11.7.2 Internally Self-Pumped Phase-Conjugate Mirror 519
11.7.3 Double Phase-Conjugate Mirror 520
11.7.4 Other Applications of Photorefractive Nonlinear Optics 521
Problems 521
References 521

Chapter 12: Optically Induced Damage and Multiphoton Absorption 523

12.1 Introduction to Optical Damage 523
12.2 Avalanche-Breakdown Model 524
12.3 Influence of Laser Pulse Duration 526
12.4 Direct Photoionization 528
12.5 Multiphoton Absorption and Multiphoton Ionization 528
12.5.1 Theory of Single- and Multiphoton Absorption and Fermi's Golden Rule 530
12.5.2 Linear (One-Photon) Absorption 532
12.5.3 Two-Photon Absorption 535
12.5.4 Multiphoton Absorption 538
Problems 538
References 538

Chapter 13: Ultrafast and Intense-Field Nonlinear Optics 541

13.1 Introduction 541
13.2 Ultrashort-Pulse Propagation Equation 541
13.3 Interpretation of the Ultrashort-Pulse Propagation Equation 547
13.3.1 Self-Steepening 548
13.3.2 Space–Time Coupling 550
13.3.3 Supercontinuum Generation 551
13.4 Intense-Field Nonlinear Optics 552
13.5 Motion of a Free Electron in a Laser Field 553
13.6 High-Harmonic Generation 555
13.7 Tunnel Ionization and the Keldysh Model 559
13.8 Nonlinear Optics of Plasmas and Relativistic Nonlinear Optics 560
13.9 Nonlinear Quantum Electrodynamics 565
Problem 567
References 567

Chapter 14: Nonlinear Optics of Plasmonic Systems *569*
14.1 Introduction to Plasmonics 569
14.2 Simple Derivation of the Plasma Frequency 569
14.3 The Drude Model 571
14.4 Optical Properties of Gold 574
14.5 Surface Plasmon Polaritons 576
14.6 Electric Field Enhancement in Plasmonic Systems 579
Problems 581
References 581

Appendices *583*
Appendix A The SI System of Units 583
A.1 Energy Relations and Poynting's Theorem 586
A.2 The Wave Equation 586
A.3 Boundary Conditions 588
Appendix B The Gaussian System of Units 590
Appendix C Systems of Units in Nonlinear Optics 594
C.1 Conversion between the Systems 595
Appendix D Relationship between Intensity and Field Strength 596
Appendix E Physical Constants 597
References 599

Index *601*

Preface to the Fourth Edition

As I was writing this Fourth Edition of my book *Nonlinear Optics*, I found the opportunity to recall the history of my intrigue with the study of nonlinear optics. I first learned about nonlinear optics during my senior year at MIT. I was taking a course in laser physics taught by Dr. Abraham Szöke. A special topic covered in the course was nonlinear optics, and Prof. Bloembergen's short book on the topic (*Nonlinear Optics*, Benjamin, 1965) was assigned as supplemental reading. I believe that it was at that point in my life that I fell in love with nonlinear optics. I am attracted to nonlinear optics for the following reasons. This topic is founded on fundamental physics including quantum mechanics and electromagnetic theory. The laboratory study of nonlinear optics involves sophisticated experimental methods. Moreover, nonlinear optics spans the disciplines of pure physics, applied physics, and engineering.

In preparing this Fourth Edition, I have corrected some typos that made their way into the Third Edition. I also tightened up and clarified the wording in many spots in the text. In addition, I added new material as follows. I added a new chapter, Chapter 14, dealing with the nonlinear optics of plasmonic systems. In Chapter 2 I added a new section on advanced phase matching concepts. These concepts include noncollinear phase matching, critical and noncritical phase matching, phase matching aspects of spontaneous parametric downconversion, the tilted pulse-front method for THz generation, and Cherenkov phase matching. The first three sections of Chapter 13 as well as Section 13.8 have been substantially rewritten to improve the pedagogical structure. A new section (Section 13.7) has been added that deals with Keldysh theory and tunneling ionization. Section 4.6 now includes a simple derivation of the Debye–Hückel screening equation. Finally, at the level of detail, I have included the following new figures: Fig. 2.3.4, Fig. 2.10.2, Fig. 5.6.2, Fig. 7.5.2, and Fig. 7.5.4.

I give my great thanks to the many students and colleagues who have made suggestions regarding the presentations given in the book and who have spotted typos and inaccuracies in

the Third Edition. My thanks go to Zahirul Alam, Aku Antikainen, Erik Bélanger, Nick Black, Frédéric Bouchard, Thomas Brabec, Steve Byrnes, Enrique Cortés-Herrera, Israel De Leon, Justin Droba, Patrick Dupre, James Emery, Marty Fejer, Alexander Gaeta, Enno Giese, Mojtaba Hajialamdari, Henry Kapteyn, Stefan Katletz, Kyung Seung Kim, Samuel Lemieux, Yanhua Lu, Svetlana Lukishova, Giulia Marcucci, Adrian Melissinos, Jean-Michel Ménard, Mohammad Mirhosseini, Margaret Murnane, Geoffrey New, Rui Qi, Markus Raschke, Razif Razali, Orad Reshef, Matthew Runyon, Akbar Safari, Mansoor Sheik-Bahae, John Sipe, Arlee Smith, Phillip Sprangle, Andrew Strikwerda, Fredrik Sy, and Anthony Vella. I also give my thanks to the many classroom students not mentioned above for their thought-provoking questions and for their overall intellectual curiosity.

Robert W. Boyd
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January 2, 2020

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.

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