## **Nonlinear Optics**

## **Nonlinear Optics**

53.74 B652

#### NICOLAAS BLOEMBERGEN

Harvard University

This book was originally published as part of the Frontiers in Physics Series, edited by David Pines.



## ADDISON-WESLEY PUBLISHING COMPANY, INC. The Advanced Book Program

Redwood City, California • Menlo Park, California • Reading, Massachusetts New York • Don Mills, Ontario • Wokingham, United Kingdom • Amsterdam Bonn • Sydney • Madrid • Singapore • Tokyo • Madrid • San Juan

#### **Nonlinear Optics**

Originally published in 1965 as part of the Frontiers in Physics Series by W.A. Benjamin, Inc.

The publisher wishes to thank the American Institute of Physics for permission to reprint material from The Physical Review.

#### Library of Congress Cataloging-in-Publication Data

Bloembergen, N.

Nonlinear optics/Nicolaas Bloembergen.

p. cm.-- (Advanced book classics)

Reprint. Originally published: Reading, Mass.: W.A. Benjamin,

1965. (Frontiers in physics; 21)

Includes bibliographical references.

1. Nonlinear optics.

I. Title II. Series.

QC446.2.B58 1991

535.2--dc20

ISBN 0-201-57868-9

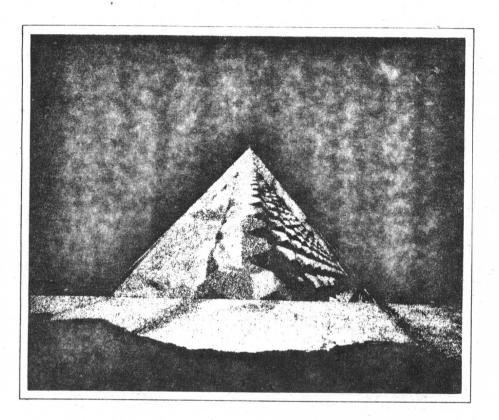
91-47920

Copyright © 1992 by Addison-Wesley Publishing, Co., Inc.

All rights reserved. No part of this publication may be reproduced, stored in a retrieval system, or transmitted, in any form or by any means, electronic, photocopying, recording, or otherwise, without the prior written permission of the publisher.

Manufactured in the United States of America. Published simultaneously in Canada.

1 2 3 4 5 6 7 8 9 10-MA-95 94 93 92 91



This unique image was created with special-effects photography. Photographs of a broken road, an office building, and a rusted object were superimposed to achieve the effect of a faceted pyramid on a futuristic plain. It originally appeared in a slide show called "Fossils of the Cyborg: From the Ancient to the Future," produced by Synapse Productions, San Francisco. Because this image evokes a fusion of classicism and dynamism, the future and the past, it was chosen as the logo for the Advanced Book Classics series.

### Publisher's Foreword

"Advanced Book Classics" is a reprint series which has come into being as a direct result of public demand for the individual volumes in this program. That was our initial criterion for launching the series. Additional criteria for selection of a book's inclusion in the series include:

- Its intrinsic value for the current scholarly buyer. It is not enough for the book to have some historic significance, but rather it must have a timeless quality attached to its content, as well. In a word, "uniqueness."
- The book's global appeal. A survey of our international markets revealed that readers of these volumes comprise a boundaryless, worldwide audience.
- The copyright date and imprint status of the book. Titles in the program are
  frequently fifteen to twenty years old. Many have gone out of print, some are
  about to go out of print. Our aim is to sustain the lifespan of these very special
  volumes.

We have devised an attractive design and trim-size for the "ABC" titles, giving the series a striking appearance, while lending the individual titles unifying identity as part of the "Advanced Book Classics" program. Since "classic" books demand a long-lasting binding, we have made them available in hardcover at an affordable price. We envision them being purchased by individuals for reference and research use, and for personal and public libraries. We also foresee their use as primary and recommended course materials for university level courses in the appropriate subject area.

The "Advanced Book Classics" program is not static. Titles will continue to be added to the series in ensuing years as works meet the criteria for inclusion which we've imposed. As the series grows, we naturally anticipate our book buying audience to grow with it. We welcome your support and your suggestions concerning future volumes in the program and invite you to communicate directly with us.

# Advanced Book Classics

P.W. Anderson, Concepts in Solids

V.I. Arnold and A. Avez, Ergodic Problems of Classical Mechanics

E. Artin and J. Tate, Class Field Theory

Michael F. Atiyah, K-Theory

Nicolaas Bloembergen, Nonlinear Optics

P.C. Clemmow and J.P. Dougherty, Electrodynamics of Particles and Plasmas

Ronald C. Davidson, Theory of Nonneutral Plasmas

P.G. deGennes, Superconductivity of Metals and Alloys

Bernard d'Espagnat, Conceptual Foundations of Quantum Mechanics, 2nd Edition

Richard Feynman, Photon-Hadron Interactions

Dieter Forster, Hydrodynamic Fluctuations, Broken Symmetry, and Correlation Functions

William Fulton, Algebraic Curves: An Introduction to Algebraic Geometry

Murray Gell-Mann and Yuval Ne'eman, The Eightfold Way

Kurt Gottfried, Quantum Mechanics

Leo Kadanoff and Gordon Baym, Quantum Statistical Mechanics

I.M. Khalatnikov, An Introduction to the Theory of Superfluidity

George W. Mackey, Unitary Group Representations in Physics, Probability and Number Theory

A. B. Migdal, Qualitative Methods in Quantum Theory

Philippe Nozières and David Pines, The Theory of Quantum Liquids, Volume II - new material

David Pines and Philippe Nozières, The Theory of Quantum Liquids, Volume 1: Normal Fermi Liquids

F. Rohrlich, Classical Charged Particles - Foundations of Their Theory

David Ruelle, Statistical Mechanics: Rigorous Results

Julian Schwinger, Particles, Sources and Fields, Volume I

Julian Schwinger, Particles, Sources and Fields, Volume II

Julian Schwinger, Particles, Sources and Fields, Volume III - new material

Julian Schwinger, Quantum Kinematics and Dynamics

Jean-Pierre Serre, Abelian & Adic Representations and Elliptic Curves

R.F. Streater and A.S. Wightman, PCT Spin and Statistics and All That

René Thom, Structural Stability and Morphogenesis

#### Vita

#### Nicolaas Bloembergen

He is Gerhard Gade University Professor, Emeritus, at Harvard University. In 1991 he serves as president of the American Physical Society. He is a recipient of the Nobel Prize for Physics in 1981, the Lorentz Medal of the Royal Dutch Academy of Sciences in 1978, the National Medal of Science, awarded by the President of the United States in 1974, the Medal of Honor of the Institute of Electrical and Electronic Engineers, and the Frederick Ives Medal of the Optical Society of America. He is a member of various Academies in the United States and abroad. In addition to his service on the faculty of Arts and Sciences at Harvard University for four decades, he was also a visiting professor in Paris, Leiden, Bangalore, Munich, Berkeley and Pasadena. Furthermore, he has served on numerous advisory committees of U.S. Government agencies and of industrial and academic institutions. He is the author of a monograph *Nuclear Magnetic Relaxation* and has published over three hundred papers in various scientific journals.

### **Special Preface**

Nonlinear Optics was written in 1964, when the field of nonlinear optics was only three years old. The available literature has since grown by at least three orders of magnitude. It is gratifying that the basic physical ideas developed in this monograph retain their validity and relevance even today. However, the reader should bear in mind that this volume gives an incomplete account. Two more recent textbooks recommended for further reading are: Y. R. Shen, Principles of Nonlinear Optics, Wiley 1985, A. Yariv, Ouantum Electronics, Third Edition, Wiley, 1988.

The vitality of *Nonlinear Optics* as a scientific discipline is illustrated by several recent developments including: soliton light pulse propagation in optical fibers for transoceanic communications, generation of femtosecond light pulses for time-resolved spectral studies and generation of "squeezed quantum states" for noise suppression. This monograph should be helpful in providing a general background of basic ideas both for experts specializing in this discipline and for scientists and students who wish to become acquainted with it.

N. Bloembergen March 1991

### **Preface**

This monograph is based on lectures, prepared for a course on quantum electronics at Harvard University in the spring of 1963 and for the summer school in Les Houches in 1964. The field of nonlinear optics is quite young. It deals with phenomena that occur at very high light intensities obtainable in laser beams. It represents one of the most interesting fields of research made possible by the development of powerful lasers.

It is perhaps foolhardy to write a monograph about nonlinear optics at this time, when new results are still announced at a high rate in scientific journals. It could be argued that such a monograph would at best contribute to its own rapid obsolescence. Nevertheless, it is hoped that it may have some more lasting value. The general principles of Maxwell's electromagnetic theory and of quantum mechanics are well established. Their domain of application is extended to include higher order interactions between light and matter in terms of nonlinear susceptibilities.

The nonlinear response of circuit elements at audio-radio and microwave frequencies is well known to the electrical engineer. In this monograph the analogous phenomena at optical frequencies are discussed. The concepts of harmonic generation, parametric amplification, modulation and rectification all have their counterparts in the visible region of the electromagnetic spectrum. The material is organized so that a pure classical description can be followed by those who have a knowledge of electromagnetic theory but are not familiar with quantum mechanics. They may skip Chapter 2 in which the quantum theory of linear and nonlinear susceptibilities is treated. This volume is intended for all who have an active interest in the field of quantum electronics, whether they are physicists interested in nonlinear electromagnetic properties of matter, electrical engineers interested in communications or high power applications at visible frequencies, or optical scientists interested in the behavior of light rays at very high intensities.

Since the field of nonlinear optics is still in a stage of rapid expansion, no effort has been made to give a complete bibliography nor to achieve a complete coverage of all

Preface

experimental data. The fundamental theoretical ideas and the basic experimental results are emphasized.

The author is indebted to Dr. J. Ducuing, Dr. Y.R. Shen, and Dr. D. Forster who have carefully read the manuscript and suggested many corrections. Any errors that remain are entirely the responsibility of the author. The permission of the edits of *The Physical Review* and the respective coauthors to reproduce the three appendices is gratefully acknowledged. The author is indebted to Drs. P.S. Pershan, and R.W. Terhune for making available some material before publication. The author wishes to express his thanks to Elizabeth Dixon who typed the entire manuscript on a tight schedule.

This monograph is dedicated to Deli Bloembergen, whose encouragement and understanding were a decisive factor in its timely completion.

N. BLOEMBERGEN

Cambridge, Massachusetts July 1964

# Preface to the Third Printing

The field of nonlinear optics has come of age. At this time the published literature in this field is at least two orders of magnitude larger than in 1964, when these lecture notes were produced. Obviously, a better and more comprehensive textbook could now be written. Lacking time and effort to accomplish that needed task, I am gratified that these original notes still provide a general framework, suitable for the description of many new developments in nonlinear optics. This reprint should satisfy an apparent demand which does not originate purely in historical curiosity. The addition of an Epilogue points out some of the shortcomings of these notes, but also shows how they provide a useful introduction to modern developments. The addition of these comments plus a selected bibliography of recent books and review papers should be helpful to the student who wishes to reach — or the more advanced worker, who is already engaged in — the current frontiers of research and engineering developments in nonlinear optics.

N. BLOEMBERGEN

Cambridge, Massachusetts Fall. 1976

# **Explanatory Notes for the Third Printing**

A bibliography covering more recent developments not included in the reprint of the main text, originally written in 1964, is added in the Epilogue of the present printing. The present book can still be used as a self-contained discussion and introduction to those fundamental principles of nonlinear optics which are describable in terms of nonlinear susceptibilities. Although no attempt has been made to update the material nor to correct all minor misprints and errors, the following notes will eliminate some difficulties which past readers have experienced in several passages.

Page 3, Eq. (1-3) and following. The definition of the amplitude adopted in this book has not survived. A definition with a factor 1/2 inserted on the right hand side of Eq. (1-3) is now in common usage. A change in definition necessitates, of course, corrections by one or more factors of two in many locations before the results here can be compared with those in other publications.

Page 5, Eq. (1-8). The complex conjugate expression should be added to the right hand side of Eq. (1-8), which should read:

$$\frac{2N_0e^3|E|^2}{m^2c(\omega^2+\bar{\tau}^2)}$$

The two sentences following this equation should be deleted.

**Page 6.** A factor 2 should be added to the right hand side of Eq. (1-2). This factor is due to the two permutations of the amplitudes  $E_1$  and  $E_2^*$  in the double product of  $(E_1 + E_2^*)^2$ . These and other permutation degeneracy factors are carefully discussed, for example, by S.K. Kurtz in *Quantum Electronics*, ed. H. Rabin and C.L. Tang, Vol. 1A, p. 209, Academic Press, New York, 1975.

Page 7. Eqs. (1-15) and (1-18) need an additional degeneracy factor of 6 on the right hand side.

Page 28. The paragraph of lines 4-20 from the top should be replaced by the following:

"The correct limiting behavior for the case that either the electromagnetic frequency or the material resonant frequency becomes very small,  $\omega \to 0$  or  $\omega_{ng} \to 0$ , respectively, requires a more careful treatment of the damping terms, as has been discussed in detail by Van Vleck and Weisskopf.<sup>3</sup>"

Page 30. A minus sign should be added to the right hand side of Eq. (2-34).

**Page 57.** " $\omega_{ab}$ " should be replaced by " $\omega_{ba}$ " on the right side of Eq. (2-98).

Page 72. In Eq. (3-26) "dE" should be replaced by "-dE".

Page 73. In Eq. (3-30) "f<sub>r</sub>" should be replaced by "f<sub>R</sub>".

Pages 121-165. The experimental results and numerical data quoted in Chapter 5 are in need of considerable revision and updating. For such information the reader is referred to the literature quoted in the Epilogue.

**Page 198.** A factor  $\in \mathbb{R}^1$  should be added to the right hand side of the second expression (Eq. 4.5).

**Page 199.** In the denominator of the last term on the right hand side of Eq. (4.12),  $\in S^{1/2}$  should be replaced by  $\in \mathbb{R}^{1/2}$ .

## **Contents**

Classical Introduction	1
Nonlinear Susceptibilities	1
Classical Atomic Models of Nonlinearity	3
The Free Electron Gas	3
The Anharmonic Oscillator	<i>3</i> 5
Magnetic Gyroscopes	8
Phenomenological Interpretation of the Nonlinear Polarization	9
Synopsis	17
References	18
Quantum Theory of Nonlinear Susceptibilities	20
The Liouville Equation for the Density Matrix	20
	. 21
	26
	31
	37
	44
Kramers-Kronig Relations	45
Quantization of the Fields	46
Nonlinear Absorption and Scattering Processes	47
	52
Coherent Quantum States, Limitation of the	
Semiclassical Treatment	55
Quantum Theory of Damping	56
References	60
	Nonlinear Susceptibilities Classical Atomic Models of Nonlinearity The Free Electron Gas The Anharmonic Oscillator Magnetic Gyroscopes Phenomenological Interpretation of the Nonlinear Polarization Synopsis References  Quantum Theory of Nonlinear Susceptibilities  The Liouville Equation for the Density Matrix Random Perturbations and Damping Response to Periodic Perturbations Lowest Order Nonlinear Conductivity Raman-Type Nonlinearities Higher Order Resonance Effects Kramers-Kronig Relations Quantization of the Fields Nonlinear Absorption and Scattering Processes Scattering Cross Sections and Nonlinear Susceptibilities Coherent Quantum States, Limitation of the Semiclassical Treatment Quantum Theory of Damping

xxi

Chapter 3	Maxwell's Equations in Nonlinear Media	•
3-1	Energy Considerations	(
3-2		ì
3-3	Coupled Wave Equations in Nonlinear Media	
3-4	A Particular Solution for Arbitrary Nonlinear Response	;
	References	7
Chapter 4	Wave Propagation in Nonlinear Media	7
4-1	Parametric Generation and Boundary Conditions Anisotropic Media	? 8
4-2	Coupling Between Two Waves: Harmonic Generation	8
4-3	Interactions with Vibrational Waves	9
. *	Acoustic Nonlinearities	9
	Brillouin Scattering	g
4-4	Parametric Down Conversion and Oscillation	g
4-5	Stimulated Raman Effect	10
4-6	Coupling Between Stokes and Antistoke's Waves	11
	References	11
Chapter 5	Experimental Results	12
5-1	Experimental Verification of the Laws of Nonlinear	
	Transmission and Reflection	12
	Geometrical Considerations	12.
	Reflected Harmonic Waves	12
	Generation of Sum and Difference Frequencies	12:
5-2	Absolute Determination of a Nonlinear Susceptibility	129
<i>5-3</i>	Multimode Structure and Fluctuation Phenomena	13.
5-4	Nonlinear Susceptibilities of Piezoelectric Crystals	134
	Temperature Dependence and Dispersion of the	
	Nonlinear Susceptibility in KDP	137
	Nonlinear Susceptibilities of Semiconductors	139
5-5	Electric Quadrupole Effects	142
5-6 5-7	Third-Harmonic Generation	144
5-7	Multiple Photon Absorption	146
5-8 5.0	Intensity Dependent Index of Refraction	147
5-9 5-10	Stimulated Raman Effect	149
5-10 5-11	Higher Order Stokes and Antistokes Radiation	153
3-11	Raman Type Susceptibilities	159
	References	163

Contents		xxii
Chapter 6	Conclusion	160
6-1	Nonlinearities in Lasers	167
6-2	Other Geometries	168
6-3	Conclusion	169
	References	169
Appendice	s	170
1	J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, "Interactions Between Light Waves in a Nonlinear Dielectric," The Physical Review, 127, 1918-1939 (1962)	171
II	N. Bloembergen and P. S. Pershan, "Light Waves at the Boundary of Nonlinear Media," The Physical Review, 128, 606-622 (1962)	193
III	N. Bloembergen and Y. R. Shen "Quantum-Theoretical Comparison of Nonlinear Susceptibilities in Parametric Media, Lasers, and Raman Lasers," The Physical Review,	
	133, A37-A49 (1964)	210
Epilogue		223
	Selected Textbooks	224
	Selected Recent Reviews	225