Fundamentals of Nonlinear Optics of Atomic Gasses

NIKOLAĬ B. DELONE
VLADIMIR P. KRAĬNOV

Fundamentals of Nonlinear Optics of Atomic Gases

NIKOLAĬ B. DELONE

Professor of Physics, General Physics Institute, Academy of Sciences of the USSR, Moscow

and

VLADIMIR P. KRAĬNOV

Professor of Physics, ZIL Autoworks Institute, Moscow, USSR

Translated from the Russian by Dr. Eugene Yankovsky

WILEY

John Wiley & Sons

New York / Chichester / Brisbane / Toronto / Singapore

***Library of Congress Cataloging in Publication Data*:**

Delone, N. B.
Fundamentals of nonlinear optics of atomic gases.
(Wiley series in pure and applied optics)
Translation of: Osnovy nelineĭnoĭ optiki atomarnykh gazov.

Bibliography: p.
Includes index.
1. Nonlinear optics. 2. Gases, Ionized gases—Optical properties. 3. Laser beams. I. Kraĭnov, V. P. (Vladimir Pavlovich), 1938– . II. Title. III. Series.

QC446.2.D4513 1987 535′.2 87-14775
ISBN 0-471-89391-9

Printed in the United States of America

10 9 8 7 6 5 4 3 2 1

Preface

Nonlinear optics came into being at the same time as the laser, about a quarter of a century ago. Since then it has become a well-developed subject of physics with many and varied applications, for it plays a definitive role in many phenomena which result when laser radiation interacts with matter.

There are many monographs devoted directly or indirectly to nonlinear optics and many of these are cited in the introduction to this book. But until now there has been no simple and consistent account of the basics of nonlinear optics that contains a description of all the interrelated phenomena, from the microscopic to the macroscopic. This, then, is the reason for this book. In it, using a simple model of a time-independent interaction of monochromatic light with an atomic gas, we describe the elementary nonlinear processes that emerge for an isolated atom, the optical characteristics of the medium averaged over a large number of atoms and depending on the intensity of the light, and the basic nonlinear optics phenomena observed in the propagation of an intense light wave through the medium.

We have focused on an analytical-theoretical description of nonlinear optics phenomena and we have illustrated conclusions with experimental results. Such an approach reflects the essence science, which is a theoretical interpretation of experimental facts and which subsequently allows one to predict unknown properties of matter. Our objective is not to describe all known nonlinear optics phenomena even within the scope of this simple model. We have, however, tried to single out the main phenomena that qualitatively distinguish nonlinear optics from the common linear optics of weak light fluxes. Although the simple model considered here ignores many aspects of nonlinear optics, the basics are presented quite fully.

In referring to the scientific literature on the subject we have given preference to monographs over reviews and reviews over original work with a view to making it easier for the reader to familiarize himself with additional material.

We have endeavored to present the material in such a way so that it is accessible to a wide range of physicists and engineers as well as to senior/graduate college students. We have assumed that the reader is familiar with the basics of atomic physics, quantum mechanics, the quantum theory of radiation, and the physics of lasers. Readers who require additional information on these subjects should turn to the *Course of Theoretical Physics* by L. D. Landau and E. M. Lifshitz (Volume 2, *The Classical Theory of Fields*, 1975, and Volume 3, *Quantum Mechanics: Nonrelativistic Theory*, 1977), to *Atomic Spectra and Radiative Transitions* by I. I. Sobelman, and to *Principles of Lasers* by O. Zvelto.

The authors are grateful to V. A. Kovarskiĭ for reviewing the manuscript and making many valuable suggestions.

NIKOLAĬ B. DELONE
VLADIMIR P. KRAĬNOV

Moscow
May 1986

Fundamentals
of Nonlinear Optics
of Atomic Gases

Notation List

LATIN SYMBOLS

a	transverse dimension of light beam
c	speed of light
$\mathbf{d}$	dipole moment
$\mathbf{E}$	electric field strength
$\mathscr{E}_n$	energy in state n
$\mathbf{e}$	polarization vector of light
$\mathbf{E}_{\mathrm{at}}$	atomic electric field strength
g	spectral-line factor
$\mathbf{H}$	magnetic field strength
I_ω	intensity of radiation of frequency ω
K	number of photons in a multiphoton process
$\mathbf{k}$	wave vector
l	orbital quantum number
m	magnetic quantum number
M_e	electron mass
n	principal quantum number
n_ω	refractive index at frequency ω
N_n	number of atoms in state n per unit volume
N_ω	number of photons of frequency ω
$\mathbf{P}$	dipole polarization vector of an atom
p	electron momentum
R	reflection coefficient of an electromagnetic wave
S	photon flux density
T, t	time of action of laser pulse
T	temperature of gas
U	atomic potential

V	interaction of radiation with atom
w	transition rate
z_{nm}	dipole matrix element

GREEK SYMBOLS

α	dipole susceptibility
Γ_n	width of state n
γ	adiabaticity parameter
Δ	resonance misfit (detuning)
$\Delta\omega$	spectral linewidth of the radiation
$\delta\mathscr{E}_n$	energy shift of state n
λ	wavelength
ν	frequency of spontaneous or stimulated radiation
ρ	density of energy levels
σ	absorption cross section
τ	natural lifetime of an atomic state
φ	phase of an elliptically polarized field
θ	angle characterizing the ellipticity of a field
$\chi^{(1)}$	linear atomic susceptibility
$\chi^{(1)}_{nn}$	linear susceptibility of an atom in state n
$\chi^{(1)}_{np}$	off-diagonal linear susceptibility
$\chi^{(K)}$	nonlinear susceptibility of order K
Ψ_n	wave function of state n
ω	frequency of incident electromagnetic wave
ω_{mn}	atomic transition frequency
$d\Omega$	solid-angle differential
Ω	Rabi frequency

Contents

Introduction

Optics is the branch of physics that studies light,—that is, the electromagnetic field in the visible range of frequencies—and its interaction with matter. The term *optics* was defined long ago and is fairly unambiguous. The term *nonlinear optics* is more ambiguous. This is quite in the nature of things, since nonlinear optics, as an independent chapter of optics, appeared rather more recently. The birth of nonlinear optics, as is well known occurred at the same time as that of the laser, in the early 1960s. Now by nonlinear optics one usually means that department of optics that studies the interaction of matter with light of high intensity, which changes the optical properties of the matter itself. If, as is customary in optics, we describe the properties of matter by the optical characteristics averaged over a large number of atoms (that is, by the polarization, dielectric constant, and index of refraction), then for light of high intensity all these characteristics become dependent on the electric field strength **E** in the light wave. For instance, the polarization vector **P** must generally be represented in the form of a series in which the terms are proportional to different powers of the electric field strength:

$$\mathbf{P} = \mathbf{P}^{(1)} + \mathbf{P}^{(2)} + \mathbf{P}^{(3)} + \cdots,$$

with $\mathbf{P}^{(1)} \propto \mathbf{E}$, $\mathbf{P}^{(2)} \propto \mathbf{E}^2$, $\mathbf{P}^{(3)} \propto \mathbf{E}^3$, etc., in the absence of resonances. Here, as everywhere in what follows, we have assumed the use of the atomic system of units ($\hbar = e = M_e = 1$), in terms of which the electric field strength E is always small compared to unity in the real situations we are interested in. Media for which such series expansions are valid and require terms beyond the first are called *nonlinear*. Allowing for the higher-order terms in the expansion of the polarization in powers of the electric field strength is equivalent to allowing for nonlinear scattering and multiphoton light absorption by the medium. For low-intensity light, that is, low electric field strength in the light field, we can ignore the higher-order terms in the expansions in comparison with the first term, which corresponds to the initial principles of ordinary optics. In accordance with this it is logical to call normal optics *linear*. This term, however, has not yet acquired wide application, although it well reflects the essence of the situation. We will be using it.

What is important is that, from the standpoint of the macroscopic phenomena that emerge in the interaction of light with matter, the difference between linear and nonlinear optics is qualitative. For instance, in nonlinear optics the basic laws of the common linear optics, such as the law of linear superposition of light fluxes and the law of rectilinear propagation of light, cease to be valid, and light of frequency ω can excite light of frequencies 2ω, 3ω, etc.

Since nonlinear optics is based on the phenomena associated with the change in the properties of matter under the action of light, it is obvious that a consistent exposition of nonlinear optics cannot be confined to a discussion of macroscopic phenomena; a microscopic discussion of the phenomena at the atomic level is also necessary. A complete and consistent exposition of nonlinear optics requires considering the elementary nonlinear effects that emerge at the atomic level and lead to the dependence of the averaged optical characteristics of matter on the intensity of light; it further requires a description of the macroscopic phenomena that appear during the propagation of a light wave in a medium, by solving the Maxwell equations and allowing for the nonlinear nature of the medium. This book is devoted to just such a thorough and consistent exposition of the basics of nonlinear optics.

One must bear in mind that the microscopic description of the interaction of light with matter was traditionally employed within the scope of the common, linear optics in such fields as molecular optics (see Born and Wolf, 1975; Fabelinskiĭ, 1968; and Vol'kenstein, 1951). Lately several monographs have been published devoted to a microscopic description of the nonlinear-optics phenomena that emerge at the atomic level (see Delone and Kraĭnov, 1985; Kovarskiĭ, 1974; and Rapoport, Zon, and Manakov, 1978). However, in classical monographs devoted to nonlinear optics—for instance, Akhmanov and Khokhlov, 1964, and Bloembergen, 1965—the focus is on the macroscopic approach and solution of the Maxwell equations for nonlinear media. Other monographs can also be cited (Apanasevich, 1977; Butylkin, Kaplan, Khronopulo, and Yakubovich, 1984; Hanna, Yuratich, and Cotter, 1979; Kielich, 1981; and Schubert and Wilhelmi, 1971) that to one degree or another consider the whole range of questions from the microscopic to the macroscopic. But the tendency of the authors of these books to describe in great detail the greatest possible number of nonlinear-optics phenomena in diverse media makes it difficult to determine the main laws of nonlinear optics.

In this book we have tried something different. We have simplified the exposition as much as possible and used a unifed language and the same approximations.

The general approach to describing the interaction is to use the common semiclassical approximation, namely, the atom is described by using quantum mechanics, while the electromagnetic field is described in terms of classical field theory. The quantum term "photon" is used only to describe the graphic transition schemes qualitatively and to clarify the meaning of energy conservation in a variable field. The classical terms "intensity of light" and "field strength in the light wave" are used equivalently, depending on the specific problem. For well-known reasons the interaction of light with an atom is described in the dipole approximation. As for the atomic transitions initiated by the light field, we assume that only one optical electron participates in such transitions.

Depending on the phenomenon, in describing the interaction of light with matter, we will be using both the language of wave optics, based on the Maxwell equations, and the language of geometrical optics, which is the classical limit of wave optics and is applicable when the dimensions of the sample are large compared with the wavelength of the light.

For the sake of simplicity we restrict our discussions to the simplest possible model of the interaction; within the scope of this model the light is assumed to be ideally monochromatic and the medium is a gas consisting of isolated atoms. It should be noted that this model is fully realistic though seemingly simple. Indeed, although laser radiation is actually quasimonochromatic, its degree of monochromaticity is very high (up to $\Delta\omega/\omega \sim 10^{-8}$). Furthermore, over a broad range of pressures, a gas can be considered as a medium consisting of isolated atoms. Finally, if we remain strictly within the optical frequency range, the differences between atomic and molecular gases are insignificant because of the small differences in the electronic spectra of atoms and molecules. Of course, in using such a model, the nonlinear-optics phenomena that occur in condensed transparent media (liquids, crystals, and glasses) are excluded in principle. However, notwithstanding the marked differences in the structure of condensed media (the presence of free electrons, the crystal lattice, anisotropy, etc.) and rarefied atomic gases, the main features of nonlinear-optics phenomena are qualitatively the same in all media. The differences are only quantitative.

Another aspect of our approach is that we deal only with steady-state processes. Processes are called *steady-state* when light acts on a substance for a much longer period than the characteristic relaxation times of the substance. When this time interval is shorter than the relaxation time, we will speak of *non-steady-state* (or *transient*) interaction. Quite naturally, such transient nonlinear-optics effects as self-induced transparency, light echo, and optical nutation (e.g. see Apanasevich, 1977) remain outside the scope of this discussion. All these effects differ qualitatively from steady-state effects. It must be noted, however, that the physical essence of these effects is related not to the nonlinearity of the medium but to the ultrashort