NONLINEAR OPTICS

E. G. Sauter

Wiley Series in Microwave and Optical Engineering
Kai Chang, Series Editor

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In memory of my parents

Preface

This textbook originated in a course of lectures in nonlinear optics that I have given for several years now at the University of Karlsruhe. Many years ago, my scientific career started with a doctoral thesis on nonlinear vibrations. I next turned my activities to high-energy physics. But now, I return to my first love: nonlinearities in optics.

The book provides an introduction to the phenomena of nonlinear optics in classical terms, avoiding quantum theory to make the book accessible to a readership not having a quantum theoretical background. This restriction forced me to discuss in some places classical phenomenological models and forbids, for example, a detailed treatment of spontaneous processes.

The background necessary for understanding this text is provided in most basic optics textbooks, some of which are listed below. The book is intended as a textbook for students in engineering and physics, as well as a reference for the practitioner who must use nonlinear optics as a new tool in his or her work. The text is written in a reasonably self-contained manner and will, hopefully, provide the student with the knowledge necessary to understand research articles on the topic.

The material can be covered in a one-semester course. The first chapter provides a general introduction to the basic relations between polarization and electric field strength. Symmetry relations help to reduce the number of independent components of the susceptibility tensors χ. A survey of the different nonlinear effects of polarizations of order two and three follows. Since we do not use quantum theory, we need a classical model that allows us to make statements on the structure of χ. This is done in the next section. The chapter ends with a discussion of the relation between the real and imaginary parts of the susceptibility.

In Chapter 2, we treat the wave propagation of complex phasors in nonlinear media and approximate the differential equations by assuming a slowly varying amplitude (or envelope). The wave propagation in anisotropic media is also treated. Conservation of energy and momentum in nonlinear processes is contained in the Manley–Rowe equations and phase matching, respectively.

In Chapter 3, the discussion of individual nonlinear effects starts with the linear electro-optic or Pockels effect (Pockels, 1893). The Faraday effect (Faraday, 1845) is an example in which polarization depends on a magnetic field. In Chapter 4, we treat second harmonic generation (Franken et al., 1961). Parametric effects are discussed in Chapter 5: sum and difference frequency mixing, frequency up and down conversion, parametric amplification, and oscillation. We begin the discussion of four-wave mixing phenomena with the Raman effect (spontaneous Raman effect: Raman, 1928; induced Raman effect: Woodbury and Ng, 1962) and the Brillouin effect (spontaneous Brillouin effect: Brillouin, 1922; stimulated Brillouin effect: Chiao et al., 1964) in Chapter 6 and continue in Chapter 7 with the quadratic electro-optic or optical Kerr effect (Kerr, 1875). As important applications, we discuss here bistability and self-focusing. In Chapter 8, the general theory of four-wave mixing is treated with phase conjugation as an application.

In Chapter 9, we investigate the propagation of light pulses first in linear media with an emphasis on pulse broadening and compression. After this, propagation in nonlinear media follows with self-phase modulation and modulation instability as applications. The important special case of solitons is discussed in Chapter 10, an old subject in principle, since J. Scott Russel first observed solitons as water waves in 1834 (described in 1844), but only in the modern laser era was attention again focused on this topic. In Chapter 11, we discuss both the harmful and useful nonlinear effects in glass fibers. While the discussions in all the other chapters will remain essentially unchanged, although some new applications may be found in the future, Chapter 11 will need revision owing to the rapid developments in the field of optical communications.

The appendices contain sections on notation and different systems of units, a table of susceptibility tensors, proof of an overall permutation symmetry, and a short description of the solution of nonlinear differential equations by inverse scattering theory.

Each chapter concludes with a collection of problems that deal with supplements to the text, analytical derivations, and applications. The text also includes numerous examples, some of which have been worked out. The reading lists at the end of each chapter contain textbooks and monographs, only in rare cases supplemented by special research articles.

Besides the omission of quantum mechanical effects, the expert will miss a discussion of spontaneous light scattering, Rayleigh scattering, photorefractive effects, nonlinear effects in semiconductors, and so on, the treatment of which would have enlarged the book too much. Some effects, as for example, nonlinear spectroscopy, are only touched upon so that the student will get at least an idea of where these effects would have their place.

In contrast to many research papers, I use here the international SI system of units. This should be convenient for students of applied physics and engineering programs. However, Appendix B gives sufficient information on how to convert these units to cgs. We choose a time dependence of the form $\exp(j\omega t)$ for stationary processes. People who prefer the negative (but you could also use "other sign") sign in the exponent, should take the complex conjugate of all functions.

In view of the great number of contributors in this area I must apologize for not being able to cite all of them in this book. I owe a great debt of thanks to Prof. G. K. Grau of the University of Karlsruhe, from whom I learned the modern aspects of nonlinear optics. His book on Quantum Electronics which unfortunately appears only in a German edition proved a steady source of knowledge for me. Without the encouragement of my friend, Prof. S. R. Seshadri of the University of Wisconsin, Madison, I would not have undertaken the task of writing this book. I am most grateful to him. Thanks are also due to my students who have helped to eliminate some of the obscurities and errors in the class notes on which this book is based. I am indebted to Mrs. D. Goldmann for her help in preparing the manuscript and to Mrs. I. Kober for drawing all the figures very carefully.

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Nonlinear Optics

Contents

CHAPTER ONE

Electric Field and Polarization

This and the following chapter constitute the basis for the treatment of special nonlinear effects in the stationary case in Chapters 3 through 8. We discuss quite generally the relation between the electric field and polarization and we define susceptibility tensors. Symmetry properties allow one to reduce the number of independent elements of the susceptibility tensors. The different specific nonlinear effects are derived up to polarization of the third order. Since we avoid quantum theory in our treatment, we have to model the nonlinear medium by a classical anharmonic oscillator to obtain the structure of the susceptibility tensors. In the linear case, the real and imaginary parts of the susceptibilities are connected by a Hilbert transform (Section 1.5), but in the nonlinear case, these dispersion relations are not very valuable. The notation is clarified in Appendix A.

1.1 THE RELATION BETWEEN E AND P; SUSCEPTIBILITY

The constitutive equation between electric displacement **D** and electric field **E** can be written in two different forms:

$$\mathbf{D} = \varepsilon \mathbf{E} = \varepsilon_0 \mathbf{E} + \mathbf{P}_L \tag{1.1}$$

In the first form, **D** is traced back to the electric field **E** using the permittivity $\varepsilon = \varepsilon_0 \varepsilon_r$; in the second form, **D** consists of a part $\varepsilon_0 \mathbf{E}$ from vacuum and a part $\mathbf{P}_L$ from matter (linear polarization). In anisotropic media, **D** and **E** are not parallel in general; ε is then a tensor of rank 2: $\varepsilon \rightarrow \boldsymbol{\varepsilon}$. Equation (1.1) solved for $\mathbf{P}_L$ leads to the definition of a first-order susceptibility tensor $\boldsymbol{\chi}^{(1)}$ (tensor of rank 2):

$$P_{Li} = \sum_{j=1}^{3} (\varepsilon_{ij} - \varepsilon_0 \delta_{ij}) E_j =: \varepsilon_0 \sum_j \chi_{ij}^{(1)} E_j \tag{1.2}$$

Here, the polarization $\mathbf{P}_L$ depends linearly on the field:

$$\mathbf{P}_L(\mathbf{E}) =: \mathbf{P}^{(1)}(\mathbf{E})$$

For strong fields, the preceding linear relation breaks down; the *most general relation* between **P** and **E** contains besides linear terms also higher powers of **E**. This leads to a nonlinear contribution to polarization:

$$\mathbf{D} = \boldsymbol{\varepsilon}\mathbf{E} + \mathbf{P}_{NL} = \varepsilon_0\mathbf{E} + \mathbf{P}_L + \mathbf{P}_{NL} = \varepsilon_0\mathbf{E} + \mathbf{P} = \boldsymbol{\varepsilon}(\mathbf{E}^0, \mathbf{E}^1, \mathbf{E}^2, \ldots) \cdot \mathbf{E} \qquad (1.3)$$

or

$$\mathbf{P} = \mathbf{P}_L + \mathbf{P}_{NL} = \mathbf{P}^{(1)}(\mathbf{E}) + \mathbf{P}^{(2)}(\mathbf{E}^2) + \mathbf{P}^{(3)}(\mathbf{E}^3) + \ldots \qquad (1.4)$$

Explicitly, the notation in Eq. (1.4) denotes the most general dependence of a time function **P** on another time function **E**, for example,

$$\mathbf{P}^{(1)}(t) = \mathbf{P}^{(1)}(\mathbf{E}^1) = \int_{-\infty}^{+\infty} d\tau\, \mathbf{q}^{(1)}(t, \tau) \cdot \mathbf{E}(\tau) \qquad (1.5)$$

$$\mathbf{P}^{(2)}(t) = \mathbf{P}^{(2)}(\mathbf{E}^2) = \int_{-\infty}^{+\infty} d\tau_1 \int_{-\infty}^{+\infty} d\tau_2\, \mathbf{q}^{(2)}(t, \tau_1, \tau_2) : \mathbf{E}(\tau_1)\mathbf{E}(\tau_2) \qquad (1.6)$$

[for the notation, see Eq. (1.10) and Appendix A.]

Remarks:

1. The field **E** is here an arbitrary function of time. Later on, Eq. (1.40), we shall decompose it into a sum of p monochromatic fields $\mathbf{E}_r$ $(r = 1, \ldots, p)$ at different frequencies. The product $\mathbf{E}(\tau_1)\mathbf{E}(\tau_2)$, for example, in Eq. (1.6) presents itself then as a sum of terms representing different processes and containing additional prefactors $c^{(n)}$ (see Section 1.3).
2. The second-order polarization $\mathbf{P}^{(2)}$ describes a *three-wave mixing* [two E fields interact and generate a third (polarization) wave], the third-order polarization $\mathbf{P}^{(3)}$ similarly *four-wave mixing* (FWM). Higher nonlinearities are of no interest here.
3 As a matter of fact, one should also consider in the expansion of **P** in Eq. (1.4) terms with magnetic field, moreover the spatial and temporal derivatives of those fields. For example, one can describe the Faraday effect with a term proportional to $\mathbf{B} \cdot \mathbf{E}$, see Section 3.4.

$\mathbf{q}(t, \tau) = (q_{ij})$ from Eq. (1.5) is a tensor of rank 2, $\mathbf{q}^{(2)}(t, \tau_1, \tau_2) = (q_{ijk})$ from Eq. (1.6) a tensor of rank 3. By the independence of physical processes from the choice of an arbitrary origin of the time scale, the form of this tensors is determined. This *independence from time displacement* (time shift invariance) means an independence of the dynamic properties of the medium from the origin of time: If, for example, the polarization $P(t_2)$ is induced by the field $E(t_1)$, then the polarization $P(t_2 + T)$ is induced by $E(t_1 + T)$.

Example 1.1 Determine $\mathbf{q}(t, \tau)$, $\mathbf{q}^{(2)}(t, \tau_1, \tau_2)$.
At first, we replace t in Eq. (1.5) by $t + T$:

$$\mathbf{P}(t+T) = \int_{-\infty}^{+\infty} d\tau\ \mathbf{q}(t+T, \tau)\mathbf{E}(\tau) \tag{1.7}$$

On the other hand, if we shift $\mathbf{E}(\tau)$ in Eq. (1.5) by T to $\mathbf{E}(\tau + T)$, we obtain due to invariance with respect to time displacement $\mathbf{P}(t) \to \mathbf{P}(t+T)$:

$$\mathbf{P}(t+T) = \int_{-\infty}^{+\infty} d\tau\ \mathbf{q}(t, \tau)\mathbf{E}(\tau+T) = \int_{-\infty}^{+\infty} d\tau\ \mathbf{q}(t, \tau - T)\mathbf{E}(\tau) \tag{1.8}$$

A comparison with Eq. (1.7) yields

$$\mathbf{q}(t+T, \tau) = \mathbf{q}(t, \tau - T) \qquad \text{for all } t, T, \tau$$

We now set $t = 0$ and then $T = t$. This results in

$$\mathbf{q}(t, \tau) = \mathbf{q}(0, \tau - t)$$

that is, $\mathbf{q}$ depends only on the difference $\tau - t$. Therefore, we write

$$\mathbf{q}(t, \tau) =: \varepsilon_0 \boldsymbol{\chi}^{(1)}(t - \tau) \tag{1.9}$$

Equation (1.6) is written in components:

$$P_i^{(2)}(t) = \sum_{j,k} \int_{-\infty}^{+\infty} d\tau_1 \int_{-\infty}^{+\infty} d\tau_2\ q_{ijk}(t, \tau_1, \tau_2) E_j(\tau_1) E_k(\tau_2) \tag{1.10}$$

If we decompose q_{ijk} into a symmetric (S_{ijk}) and an antisymmetric (A_{ijk}) part with respect to a permutation of the pairs (j, τ_1) and (k, τ_2), we see that the antisymmetric part yields a vanishing contribution after multiplication with $E_j(\tau_1)E_k(\tau_2)$ in Eq. (1.10). So we can assume $A_{ijk} = 0$ without restricting generality (see Problem 1.2) and take q as symmetric in the pairs (j, τ_1), (k, τ_2):

$$q_{ijk}(t, \tau_1, \tau_2) = q_{ikj}(t, \tau_2, \tau_1) \tag{1.11}$$