QUANTUM ELECTRONICS: A TREATISE

HERBERT RABIN
C. L. TANG

VOLUME I
Nonlinear Optics,
Part A

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Nonlinear Optics, Part A

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VOLUME I

Nonlinear Optics, Part A

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Preface

Following the advent of the laser, quantum electronics has emerged as a multidisciplinary subject of great breadth and richness, attracting the interests of basic as well as applied research workers. This subject, accordingly, is characterized by numerous specialized research reported in a wide range of original literature sources. In this treatise, "Quantum Electronics," through review articles written by principal workers in their respective fields, it is planned to present unified discussions of major quantum electronics topics. Through this approach it is hoped to stimulate understanding and progress in quantum electronics by making it relatively easy for an advanced student or investigators with limited prior background to survey topics of interest, as well as by providing reviews containing original material, both in content and organizational style, of benefit to advanced workers in the field.

Volumes IA and IB deal with the topic of nonlinear optics, with review articles on the subjects of nonlinear optical susceptibilities, nonlinear optical processes, and applications. The topics selected in these first volumes represent a compromise between the large number of subjects now embodied by nonlinear optics, an attempt to represent a balance between theoretical and experimental interests, and, of course, the general availability of authors. We are indebted to our colleagues, the authors, for their contributions.

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General Introduction

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I. HISTORICAL BACKGROUND

The field of nonlinear optics is concerned with physical phenomena that are based on a nonlinear response of a medium to applied electromagnetic fields, with at least some of the frequencies lying in the infrared, visible, ultraviolet, or x-ray region of the spectrum. The electric polarization or the induced electric current density may, for example, be a quadratic, cubic, or even an exponential function of the electric field amplitudes. Alternatively, one may say that the dielectric and magnetic susceptibilities, or the optical index of refraction, are themselves functions of the field strengths. With this general definition, the field of nonlinear optics has a venerable history.

Maxwell's equations remain valid if the constitutive relationships between **D**, **E**, **B**, and **H** become nonlinear. One of the first and best-known examples is the nonlinear relationship $\mathbf{B} = \mu(\mathbf{H})\mathbf{H}$ between magnetic induction and field in ferromagnetic materials. Since generators, transformers, and inductors usually contain ferromagnetics, harmonic distortion of electromagnetic waves occurs in power circuits at 50–60 Hz. Such distortion is especially harmful in audio equipment. There are, of course, no optical frequencies involved in this particular example, but nonlinearities in the constitutive relations in the optical region were also known in the 19th century. The linear electrooptic, or Pockels, effect and the quadratic electrooptic, or Kerr, effect represent situations [see, e.g., Pockels (1906); cf. Szivessy (1929)] in which the index of refraction, or dielectric constant or susceptibility, is itself a linear or quadratic function of the applied electric field. They may be described by the following nonlinear constitutive relations:

$$P_i = \chi_{ijk}^{(2)} E_j E_k \tag{1}$$

$$P_i = \chi_{ijkl}^{(3)} E_j E_k E_l \tag{2}$$

For the Pockels effect one of the electric field components appearing on the right-hand side of Eq. (1) is taken at an optical frequency and the other component at zero frequency. In the same manner the Kerr effect is contained in Eq. (2), in combination with the other Maxwell equations, if one takes for two field components on the right side the applied dc electric field.

The phenomenological constitutive relationships, as expressed by Eqs. (1) and (2), remain formally valid and exist for arbitrary frequency components on the right-hand side. Just as the linear susceptibility $\chi_{ij}^{(1)}(\omega)$ is a function of frequency in dispersive media, so are the nonlinear susceptibilities functions of the various frequencies of the electric field components [see e.g., Bloembergen (1965)]. If the applied dc electric field in the Pockels effect is replaced by a second wave at an optical frequency, it is easy to see that the quadratic relationship between electric polarization and applied fields leads to responses characteristic of any quadratic device. The nonlinear susceptibility $\chi^{(2)}$ thus implies the following optical processes:

(a) second harmonic generation of light,
(b) rectification of light,
(c) sum frequency generation, and
(d) difference frequency generation.

Similar phenomena have, of course, long been known in the radio and microwave region of the electromagnetic spectrum. They have found widespread application in devices, such as diode detectors, modulators, demodulators, harmonic and parametric generators, etc. Such devices have also been constructed at optical frequencies, as discussed in the present volume. An essential feature of nonlinear optics is that physical dimensions of the material are usually large compared to the optical wavelength. Therefore wave propagation characteristics are very important.

The Faraday and Cotton–Mouton effects (Szivessy, 1929) are examples of a situation in which the optical index of refraction is a linear or quadratic function, respectively, of an applied magnetic field. Again the magnetic field components may be varying in time. Since the time derivatives of magnetic field components are related to spatial derivatives of the electric field components by Maxwell's equations, and since the electric current density is related to the polarization by $j = \partial \mathbf{P}/\partial t$, the general nonlinear constitutive relationship may be written in terms of nonlinear conductivities:

$$j_i = \sigma_{i,jk\ldots,opq\ldots}(\nabla_j \nabla_k \cdots) E_o E_p E_q \cdots \tag{3}$$

For most situations one need not go further than conductivities with three or four indices, described by third or fourth rank tensors. Some highly nonlinear effects have, however, an exponential or transcendental character. In such cases the power series expansions in Eqs. (1)–(3) are not a good approximation. A very old example, known long before Maxwell's time, of an exponential nonlinearity is the phenomenon of electric breakdown in gases and condensed matter. Laser beams can also cause electric breakdown and this is a highly nonlinear optical effect.

The photoelectric effect, either the internal photoconductive type or the external photoemission, is an example of rectification by a quadratic response, although it often is not considered as a part of nonlinear optics. The photoelectric current $j_{dc} = CEE^*$ is proportional to the absolute square of the field amplitude. Forrester *et al.* (1955) considered the demodulation by the photoelectric effect when the incident light field contains Fourier components at ω_1 and ω_2, respectively, and calculated the Fourier component at the difference frequency $\omega_1 - \omega_2$ in the photocurrent.

A nonlinear response such as given by Eqs. (1) and (2) may be obtained from a simple classical model. A valence electron is regarded as a slightly anharmonic oscillator. The strictly harmonic oscillator model introduced by Lorentz (1909) to explain the linear polarization does not give rise to nonlinearities. If the restoring force is not strictly proportional to the deviation from the equilibrium position, the nonlinear response terms appear. Since the valence electrons are bound in the Coulomb field of the ion core, the restoring forces are certainly not proportional to sizeable deviations. The energy levels of the electron are not equidistant, as they would be for a harmonic oscillator. Thus all materials have nonlinear electromagnetic properties, which should become evident at sufficiently high values of the applied fields.

The response functions to electromagnetic fields may be calculated quantum mechanically by higher-order, time-dependent, perturbation theory. An early example of this is the calculation of two-photon absorption processes by Goeppert-Mayer (1931). Even the vacuum has nonlinear properties if the virtual creation of electron–positron pairs is considered. The nonlinear susceptibility of the vacuum is discussed in textbooks on quantum electrodynamics (Akhiezer and Beresteskii 1965) and was introduced by Heisenberg [see Heisenberg and Euler (1936)] in 1936.

The basic principles of electromagnetic and quantum theory were firmly established and available as a foundation for the development of nonlinear optics in the early 1930s. Nevertheless the field did not develop as a well-defined scientific discipline until the 1960s because the striking phenomena of nonlinear optics can readily be observed only at high power density levels. A relevant parameter is the ratio of the electric field amplitude in the light wave to the electric field responsible for the binding of the electron inside the atoms, molecules, or crystal lattices. The atomic Coulomb field is on the order of 3×10^8 V/cm and nonlinear phenomena are either very difficult or impossible[1] to observe if light field amplitudes are less than 300 V/cm corresponding to power flux densities of 250 W/cm^2. With the advent of lasers much higher power flux densities became available. As soon as the first powerful ruby laser was developed, Franken and co-workers (1961) observed the second harmonic generation in the ultraviolet at 3470 Å, when the ruby laser with $\lambda = 6940$ Å wavelength traverses a quartz crystal.

During the past decade the field has rapidly developed into an active scientific discipline. The same concepts that were already familiar at low frequencies, i.e., in the audio, radio and microwave region of the spectrum, could readily be extended to the optical region (Bloembergen, 1965). Many new phenomena, such as optical parametric up and down conversion, stimulated Raman and Brillouin scattering, self-focusing of light beams, self-induced transparency, to name just a few, were discovered. At the same time, some older phenomena, which were mentioned earlier in the introduction, could be understood in a broader and more general framework. The field of nonlinear optics has become an integral part of textbooks on lasers and quantum electronics (Yariv, 1967; Pantell and Puthoff, 1969; Marcuse, 1970). The operation of lasers and many of their applications are based in an essential manner on nonlinearities. Even the field of holography falls within the general definition of nonlinear optics given in this section. In making a hologram one produces a change in the real or imaginary part of the index of refraction of the recording material, which is proportional to the product of the optical field from the object and the reference beam. In view of the pervasive nature of these nonlinear responses, it is entirely appropriate and timely to publish this volume on nonlinear optics.

II. THE PRESENT VOLUME

The second chapter in Part I deals appropriately with a discussion of the nonlinear susceptibilities, i.e., the coefficients appearing in the nonlinear constitutive relationships. They may be calculated quantum mechanically for

[1] An exception is provided by nonlinear phenomena near strong and narrow resonances, which occur, for example, in nonlinear saturation spectroscopy in gases (Brewer, 1972).

different classes of materials. Their experimental determination and numerical values for a variety of materials are the subject of the third chapter of Part I.

Part II of this volume describes several nonlinear optical phenomena to which these nonlinear susceptibilities give rise. The characteristics of these basic nonlinear interactions are presented in a unified and cohesive fashion. The self-contained treatment should, however, not cause the reader to think that the subject matter of nonlinear optics is exhausted and closed by this one volume.

The subjects of laser-induced electric breakdown, self-induced transparency, photoelectric mixing and, holography could not be treated in this volume. Stimulated scattering from concentration and temperature fluctuations, plasma waves, spin waves, and other elementary excitations are not discussed in detail. Self-focusing, saturation, and other consequences of an intensity dependent index of refraction are mentioned only in so far as they relate directly to the main subject of each chapter. The field of high-resolution saturation spectroscopy in gases will probably need a volume of its own (Brewer, 1972). One could wish for a separate chapter on the optical laws of reflection, refraction, and diffraction in the nonlinear case (Bloembergen, 1965). There exist nonlinear analogs of total reflection, Brewster's angle, internal and external conical refraction in biaxial crystals, to name just a few of the topics that may be found in linear optics textbooks. Although it is impossible to achieve completeness in these two volumes, the reader will catch the spirit of nonlinear optics from the up-to-date account of the topics mentioned in the chapter headings.

In the same manner the chapters in Volume IB, Part III cover several important applications in depth, rather than give an encyclopedic survey of all nonlinear optical devices. Modulators, demodulators, mode-locking devices, thin film optical switches, and many other applications exist. The applications treated in detail in the present volumes are those concerned with the generation of coherent light at new frequencies.

III. FUTURE OUTLOOK

If an extrapolation is made from the experimental origin of nonlinear optics in 1961 through the past decade and the present level of activity into the future, one may reasonably expect that many new results will be obtained in the next decade. The science of nonlinear spectroscopy has only just begun, as coherent light beams, tunable over a wide spectral range, have recently become available through the development of dye lasers and parametric oscillators. High-resolution, nonlinear, absorption spectroscopy of molecules in the ultraviolet and far infrared will require much effort. The nonlinear spectroscopy of excitations in crytals will augment our fragmentary knowledge of the frequency dependence of nonlinear susceptiblities. The future availability of high intensity x-ray sources, from synchrotron radiation and perhaps from x-ray lasers, will enable the extension of nonlinear spectroscopy beyond the ultraviolet. It should be realized that at this time our knowledge of nonlinear properties even in the near ultraviolet is very scant indeed. Many important applications in photochemistry appear on the horizon. Very short lifetimes of molecular excitations in fluids can now be measured and extension of transient spectroscopic analysis to the subpicosecond regime appears feasible. Precise analysis of nonlinear plasma phenomena will be of importance in the current efforts to achieve laser-produced thermonuclear plasmas. Understanding of nonlinear properties is essential for the design of components and handling of high-power laser beams. Considering the continued activity and vitality of the venerable science of linear optics, one may expect with some confidence a rather long-lived and fruitful activity in the field of nonlinear optics and nonlinear spectroscopy. Research workers of the coming decade will find up-to-date reviews of integral parts of nonlinear optics, as embodied in this volume, useful in making further progress. They owe a debt of gratitude to the editors and to the authors, who have been willing to take time from their distinguished research activities to produce the present volumes.

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Part I

NONLINEAR OPTICAL SUSCEPTIBILITIES