Book Review

Nonlinear Optics, Third Edition

Robert W. Boyd, 613 pages +xix, ISBN: 978-0-12-369470-6, illus., index, Academic Press, San Diego (2008), $99.95 hardcover.

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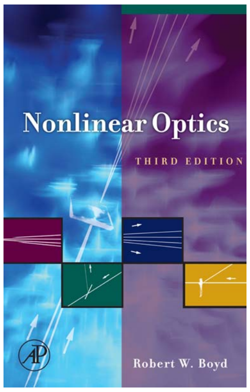

Nonlinear optics is emerging as an important imaging technique within the scope of biomedical optics. For example, the readers of the *Journal of Biomedical Optics* (JBO) are familiar with the many excellent papers that are published in the journal on the topic of nonlinear optics. There exist several comprehensive textbooks on nonlinear optics; however, I highly recommend the third edition of *Nonlinear Optics* as a reference book. Its use as a textbook for a graduate course in physics would require a truncated syllabus that covers selected chapters from the book.

First, it is important to define clearly both linear and nonlinear optics as the author does on the second page of his book. Linear optical phenomena are characterized by a induced polarization $P(t)$ in the material that depends in a linear manner on the electric field strength: $E(t)$: $P(t)=\varepsilon_0\chi^{(1)}E(t)$. The constant of proportionality $\chi^{(1)}$ is known as the linear susceptibility, and ε_0 is the permittivity of free space. For the case of nonlinear optics, the optical response is described by expressing the induced polarization $P(t)$ as a power series of the electric field strength:

$$P(t) = \varepsilon_0[\chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + \dots].$$

The quantities $\chi^{(2)}$ and $\chi^{(3)}$ are known as the second- and third-order nonlinear optical susceptibilities. In writing these two equations, an important assumption is made: the polarization of the material at time t only depends on the instantaneous value of the electric field strength. It follows from this assumption, as Boyd points out through the Kramers-Kronig relations, that the medium must be lossless and dispersionless. The author also shows how to modify and extend the treatment for materials with loss and dispersion. Typically, nonlinear optical phenomena occur in the presence of laser-matter interactions in the presence of very high electric field strengths.

The author's intended audience for *Nonlinear Optics* consists of advanced undergraduates and graduate students who are working in the fields of optics, physics, chemistry, electrical engineering, mechanical engineering, and chemical engineering. Since the book contains more material than can be covered in a single course, in his preface the author provides some advice to instructors who may select his book as a textbook (e.g., which chapters are critical and which chapters can be deleted from a course syllabus). As a textbook, the wide range of problems at the end of each chapter contributes to the reader's understanding of the material. A well-designed index is provided.

In order to ascertain the reader's level of understanding of nonlinear optics, and therefore the suitability of *Nonlinear Optics* as a reference, I pose the following problem: please explain the physical and the mathematical basis of the statement that second-harmonic generation will not occur in materials that have centrosymmetry or the presence of a center of inversion. Such materials include isotropic materials such as liquids, glasses, and certain types of crystals, such as cubic crystals. For these materials the lowest order of nonlinear optical susceptibility is the third-order susceptibility. The explanation should include a statement of both the physical phenomenon and the mathematical analysis that is the basis of the previous statement. If you cannot readily show the physical and mathematical basis of the statement, then please consult a reference book on nonlinear optics. Is it still not very clear? Then I suggest you read Chap. 1, pp. 44–52 of Boyd's *Nonlinear Optics*. Initially, Boyd first states that his treatment is similar to or closely follows that of another author, and he gives the complete citation, which in this case is Owyoung (1971). Boyd concisely proves mathematically that the second-order nonlinear optical susceptibility vanishes for crystals with an inversion symmetry. Then he proceeds to give a clear physical explanation of why this occurs.

This illustrative example shows three aspects of what the reader can expect to find in Boyd's clear, concise, and properly attributed expositions with ancillary graphic illustrations of nonlinear optical phenomenon. First, the author clearly cites the proper attribution when he follows a previously published treatment. Second, the author provides a rigorous mathematical analysis of the physical phenomenon. The author used the International System of Units (SI) throughout the book. He begins the derivation for the case of limited physical and mathematical assumptions and approximations (all of which are defined and stated) and moves to mathematical expressions that are simplified through the use of assumptions and approximations. Third, the author provides clear and physically reasonable explanations for the phenomena that are consistent with their mathematical descriptions. Finally, the author refers to published papers that contain physical measurements of the phenomena. In that way he compares the theoretical values with the experimentally measured quantities in order to test and validate the theory.

I now describe some of the unique and eminently useful features of the third edition of *Nonlinear Optics*. Since the second edition is on many book shelves and widely used, it is always a legitimate question to ask what is new and different in the third edition. The previous editions were written using the Gaussian system of units, while the third edition has been entirely rewritten using the SI (or rationalized MKSA) system of units. MKSA includes the units of meter, kilogram, seconds, and ampere. The new third edition provides expanded appendices that discuss the SI and Gaussian systems of units and procedures for their conversion, as well as tables that give the relationship between intensity and field strength, a table of physical constants for both the Gaussian system with the centimeter, gram, and second (CGS) units and the SI systems, physical constants that are specific to the SI system, and a table of conversion of units between the systems. Other major additions include sections on the applications of harmonic generation to microscopy and biophotonics, and a section on spectroscopy that is based on coherent anti-Stokes Raman scattering.

At the end of each chapter there is a reference section that includes several subsections: reviews on the topics covered in the chapter, further readings on the topic that are typically monographs, a section on applications of the topics, and a selection of key peer-reviewed publications. All of the experimental data given in the chapters and the details of the instrumentation from which they were measured are fully cited in the references. I strongly recommend that the reader peruse the cited primary literature.

The reader will not find this book to be a useful reference for the detailed experimental aspects of nonlinear optics. In particular, this book is not an appropriate source and guide for the reader who wishes to construct instrumentation that is intended to measure nonlinear optical phenomenon. Alternatively, the references contain these experimental details.

The astute reader may ask the following question: why are there various sets of units—the Gaussian (a variant of CGS) and the SI systems of units? Two short answers are history and convenience. Note the systems of units in both mechanics and in electromagnetism are arbitrary. In 1833 Carl Friedrich Gauss proposed the metric system (millimeter, gram, and second) and in 1874 it was extended by James Clerk Maxwell and William Thompson to a set of electromagnetic units. In 1874 the British Association for the Advancement of Science introduced the CGS system that was later replaced with the more practical MKSA, which was then developed into the current SI system of units.

As Jackson points out in his comprehensive appendix on units and dimensions in his third edition of *Classical Electrodynamics*, both convenience and clarity are desirable features of a system of units. While in the field of mechanics there is some consensus with the units of length (centimeter or meter), mass (gram or kilogram), and time (mean solar second), in electromagnetism the situation is more complicated. Historical varieties of CGS units for electromagnetic theory include electromagnetic units (emu), Gaussian units, and Heaviside-Lorentz units.

Different authors use different systems of units; for example, CGS units are used in some astrophysical journals. Additionally, there are theoretical physicists who use a system of natural units in which all physical quantities are measured in terms of mass (or electron volts) as the only basic unit. All other quantities, such as length, time, force, or energy, are expressed in terms of this one unit and written as powers of this dimension.

The third edition of *Nonlinear Optics* uses the SI system of units, so let us look at that system first. In the SI system of units the vacuum is treated as a material medium of ε_0 permittivity and permeability μ_0. Therefore, the factors ε_0 and μ_0 appear in the equations. Also there is the factor of 4π. In the SI system, distance is measured in meters (m), mass in kilograms (kg), and time in seconds (s). The meter is defined as the distance that light travels in a given interval of time. The second is defined by 9,192,631,770 oscillations between the two hyperfine levels of the ground state of a ^{133}Cs atom at rest at a temperature of 0 K. The newton (N) is the unit of force, which is kg m/s^2. The joule (J) is the unit of energy, which is kg m^2/s^2. The coulomb (C) is the unit of electrical charge, defined such that the force between two charged particles, each of 1 C of charge and separated by 1 m, is 1 N. The unit of electrical current is the ampere (A), which is 1 C/s. Finally, the volt is the unit of electrical potential, which is 1 J/C. With these definitions, the author wrote the Maxwell equations using the SI units of measure. The units and the names of the field vectors are then given: electric field, electric displacement, magnetic field or induction, magnetic intensity, polarization, and magnetization. The constitutive relations give the relations between the electromagnetic field vectors and the properties of materials. The author derives Poynting's theorem (the Poynting vector is the rate at which electromagnetic energy passes across a unit area that is normal to the direction of propagation) from Maxwell's equations.

Next the author derives the wave equation from Maxwell's equations. This is of fundamental physical importance, because under the assumption that the electromagnetic wave is propagating in a linear, isotropic, nonmagnetic medium that is free of sources, the electric and magnetic fields and the wave vector are all mutually orthogonal. It also follows for propagation of the electromagnetic wave in a vacuum that its speed is equal to the speed of light.

In the next appendix the author presents the equations of electromagnetism as they are written in the Gaussian system of units (a variant of the CGS system). For the mechanical properties, distance is measured in centimeters (cm), mass is measured in grams (g), and time is measured in seconds (s). While the CGS system is the same for mechanical units, there are several variants for electromagnetism. Force is given in units of g cm/s^2 (dyne) and energy is given in units of g cm^2/s^2 (erg). Charge is defined as the charge of a given number of electrons and is called the statcoulomb or the electrostatic unit of charge. This is defined such that the force between two charged particles, each containing 1 statcolumb of charge and separated by 1 cm, is 1 dyn. Similarly, the unit of current is the statcoulomb/s, which is called the statampere.

The unit of electrical potential is the erg/statcoulomb, known at the statvolt.

As the author cogently points out, in the Gaussian system of units the four primary field vectors (the electric field, the electric displacement field, the magnetic induction, and the magnetic intensity), as well as the polarization vector and the magnetization vector, all have the same dimensions! The common dimension is the statvolt/cm or the statcoulomb/cm^2, which are also called the gauss (G) or the oersted (Oe). Typically, the unit of gauss only refers to the magnetic field, and the unit of oersted only refers to the magnetic intensity. It should be pointed out that in the SI system and in the CGS systems, the units differ in their dimensions.

As with the SI system of units, the author derives Poynting's theorem and the wave equation. These and the other derivations in the book are clear and easy to follow. From Maxwell's equations it follows mathematically that the electric and magnetic fields and the direction of energy propagation are all mutually orthogonal. In the last appendix the author presents the units of nonlinear optics in the two types of systems of units, shows how to convert between these two systems, and provides tables of physical constants in these two systems. These appendices are extremely useful, and it is rare (see Jackson's third edition) to find such detailed discussion of these two systems of units in a nonlinear optics textbook.

So why do some authors prefer the CGS system of units, and others prefer the Gaussian system of units? Perhaps the CGS system simplifies their theoretical calculations; on the other hand, in electromagnetism the CGS units are difficult to define experimentally. The latter problem is not the case for the SI system of units since it is based on the ampere, a unit of current that is easily defined experimentally, with measurements of the quantum Hall or the Josephson effects, for example. However, the constants in the electromagnetic equations are more complex.

The book begins with the definition of linear and nonlinear susceptibilities for material that is lossless and dispersionless. Next the author presents the more general case with dispersion and loss. Starting with the Lorentz model of an atom as a harmonic oscillator, which is a good description of the linear optical properties of atoms, he proceeds with the classical anharmonic oscillator. This treatment clearly shows that for the case of a material without a center of inversion there is a second-order optical nonlinearity. However, for the case of a material with a center of inversion, the lowest order of nonlinearity is the third-order nonlinear susceptibility. The author cogently points out the major limitation of the classical model of optical nonlinearities: it assumes a single resonance frequency for each atom. The application of quantum mechanics to the theory of nonlinear and linear optical susceptibilities solves this severe limitation in that each atom can have many energy eigenvalues, and thus several resonance frequencies.

Often in other books one reads about the Kramers-Kronig relations in linear and nonlinear optics (they are always valid in linear optics, and sometimes valid in nonlinear optics); however, a physical interpretation and a rigorous derivation are usually lacking. That is not the case in Boyd's book. Boyd explains that these relations relate the real and the imaginary parts of the frequency-dependent quantities such as linear susceptibility. They are extremely useful since they permit the real part of the susceptibility at a specific frequency to be determined from the frequency dependence of the imaginary part of the susceptibility. It is easier to measure the absorption spectrum than to determine the frequency dependence of the refractive index. By treating the frequency as a complex quantity and by using the technique of complex integration, the author develops a clear derivation of the Kramers-Kronig relations.

Following the classical treatment of nonlinear optical susceptibility with its strong pedagogical value, the author demonstrates how quantum mechanics can be used to derive the nonlinear optical susceptibility. A prerequisite for understanding this material is to have a good grounding in quantum mechanics; however, the rewards of this approach include the following. The expressions show the symmetries of the susceptibility and how the susceptibility depends on the dipole transition moments and the atomic energy levels, and the expressions can predict the numerical values of the nonlinear optical susceptibilities. Boyd clearly shows how the use of quantum mechanical perturbation theory to solve Schrödinger's equation yields expressions for the linear, the second-order, and the third-order susceptibility. The author then provides a clear derivation of the use of density matrix calculations (an alternative form of quantum mechanics) to yield the same quantities.

Nonlinear Optics is both clear in its exposition and comprehensive in its treatment of the field. As two final exemplars of this characterization I select two topics that may be of special interest to the readers of JBO: optical phase conjugation and multiphoton processes.

In 1985 Zel'dovich et al. showed that optical phase conjugation can be used to remove or cancel the effects of optical aberrations in some types of optical systems. The phenomenon is sometimes called wavefront reversal, but actually the generated wavefront is the same as the incident wavefront, but it propagates in the opposite direction. The author first qualitatively describes the effect of a source or signal wave that is reflected from a perfect or ideal phase conjugate mirror to produce a reflected or phase-conjugate wave with the following properties. First, the complex polarization unit vector of the incident wave is replaced by its complex conjugate. Second, the amplitude of the incident wave is replaced by its complex conjugate. Third, the wave vector of the incident wave is replaced by its negative, which is equivalent to time t being replaced by its negative, or $-t$ (time reversal). Following this qualitative description of the process, the author demonstrates mathematically how optical phase conjugation leads to aberration correction. Additionally, the author derives the mathematical relations that show how phase conjugation can be generated by the nonlinear optical process of degenerate four-wave mixing. Again, this section is an excellent example of presenting both the physics and its mathematical description. The perceptive reader will immediately realize that there are interesting applications of phase conjugation to microscopy.

The final exemplar is a very concise section on multiphoton absorption and multiphoton ionization. I found the author's quantum mechanical derivation of single- and multiphoton absorption cross sections based on Fermi's golden rule and the rotating wave approximation to be especially clear. The author ends the section by generalizing the linear and two-photon absorption processes to higher order processes and presenting the transition rates for these processes.

While I have only described some of the topics that are elegantly covered in *Nonlinear Optics, Third Edition* there is a wealth of other topics that are clearly described in the book, such as phase matching, spontaneous light scattering and acousto-optics, stimulated Brillouin, Rayleigh, and Raman scattering, and ultrafast and intense-field nonlinear optics. The value of this highly recommended book is its ability to explain complex phenomena, and at the same time to provide the underlying rigorous mathematical descriptions.

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