NONLINEAR OPTICS

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Advanced Topics in the Interdisciplinary Mathematical Sciences

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PREFACE

THE GOAL OF THE ATIMS series, of which this is the first monograph, is to provide texts in areas of intense current interest for graduate students and newcomers to the field, which will take the reader from the level of first-year graduate coursework to some of the frontiers of modern research. It is intended that the monographs be written by active researchers in the field and that they give a clear introduction to, and a unified picture of, the subject. No monograph is intended to be a tome, the final word. Such a goal is impossible in rapidly moving fields for two reasons. First, researchers simply do not have the time and, even if they did, the final word would no longer enjoy that status one year later. ATIMS therefore has introduced a novel concept. Monographs that prove to be popular will be updated at regular and appropriate intervals (e.g., three years). In this way, the successful monograph will evolve into a definitive treatise.

The first monograph in the series is about Nonlinear Optics, the study of how high-intensity light propagates through and interacts with matter. It is a subject so scientifically rich and technologically promising that it is destined to become one of the most important areas of scientific research over the next quarter-century. The book is written for graduate students and the newcomer to nonlinear optics, or anyone who wants to get a unified picture of the whole subject. It takes the reader from the starting point of Maxwell's equations to the frontiers of modern research in the subject.

The modeling—how one pictures light and matter—and the mathematical methods are explained clearly and in great detail. In particular, the book starts from the point of view that light and matter each can be viewed as systems of oscillators, and the coupling between these oscillators is relatively weak. Essentially, the oscillators of light are plane electromagnetic waves and those of matter are electronic transitions, molecular vibrations and rotations, and acoustic waves. In semiconductors, which are not covered in this first edition, they are Bloch wavefunctions. Because of weak coupling, we show how all the fast time (10^{-15} second) and space (10^{-6} meter) scales can be removed from the description of the interactions and how one is then left with well-known universal and canonical equations such as the coupled mode equations (useful for switches and in situations where linear and nonlinear birefringent effects are important), the nonlinear Schrödinger equation (useful for fiber optics and nonlinear waveguides), the three- and four-wave interaction equations (useful for understanding Raman and Brillouin scattering and phase conjugation), the Maxwell–Bloch equations (useful for lasers and understanding optical bistability), the Maxwell–Debye equations (for describing delayed response in transparent materials), the sine–Gordon equation (useful for describing the propagation of light pulses through active media), and the complex Ginzburg–Landau equation (useful near the onset of lasting action).

The material is best taught in a year-long course. Students should have some familiarity with mathematical methods, Fourier series and Fourier integrals, elementary complex variables, elementary ordinary and partial differential equations, and vector calculus and should have had a course (undergraduate level is sufficient) in electromagnetic theory. Chapter 6, "Mathematical and Computational Methods," should be read concurrently with many of the sections in the other chapters. The correspondence between this chapter and the other chapters is indicated by the number contained in square brackets after the listing of each section in the table of contents. Important cross-reference sections are also indicated after the section headings in the text proper. A one semester course might cover Chapter 2, Chapter 3, Sections a, c–g, Chapter 4, Sections a–d, Chapter 5, Section a, b together with the relevant material from Chapter 6 including the uses of numerical simulations.

We begin in Chapter 2 with the propagation of light beams in weakly nonlinear (Kerr) media and, using elementary mathematics and heuristic arguments, introduce the reader to the notion of linear and nonlinear refractive index, intensity-dependent phase modulation, wavepackets, dispersion, diffraction and the nonlinear Schrödinger (NLS) equation. This is followed by a discussion of linear and nonlinear birefringence and three- and four-wave mixing. By the end of the first chapter, the reader will have met some of the most important equations describing propagation in passive nonlinear media and encountered some of their most important properties. Chapter 3 discusses communication in optical fibers using nonlinear pulses (solitons) as information bits and the reflection and transmission characteristics (Snell's or Descartes' laws) of light beams at the interfaces of different nonlinear dielectrics. To this point, matter has played a more or less passive role. The effects of delay and field intensity are included through the use of a frequency- and intensity-dependent refractive index. In Chapter 4, the oscillations of matter become active field variables, and we derive the Maxwell–Bloch and

Maxwell–Debye equations from first principles. The basic mechanisms of Raman and Brillouin scattering are introduced. Chapter 5 is about applications, lasers, optically bistable cavities, co- and counterpropagating beam interactions, coherent pulse propagation in inhomogenously broadened media, and finally Raman and Brillouin scattering.

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Chapter 1

INTRODUCTION

General Discussion

MAXWELL'S THEORY OF ELECTRIC AND magnetic fields and his idea that light is an electromagnetic wave were some of the great milestones of scientific thought, and unified understanding of a large and diverse set of phenomena. Indeed, by the late nineteenth century, the success of the classical electromagnetic theory of light led some to believe that there were few new fundamental discoveries to be made. This smug complacency was soon shattered by the inability of the wave theory to explain several observations: radiation spectra, the photoelectric effect, x-rays, radioactivity. These effects could only be understood by reviving the idea of the corpuscular nature of light, not in the original form conceived by Newton, but in a way that was compatible with the considerable success enjoyed by the classical wave theory. Out of this effort, modern quantum theory was born, and optical science settled once again into the complacency of a solved science. Rapid progress was made. The accuracy of the geometrical optics approximation, together with the linearity of the equations, which meant that complicated solution fields could be built by the linear superposition of much simpler solutions (e.g., plane waves), played important roles in this development. The amplitude of the electromagnetic field seemed to matter little.

There were, of course, rumblings, suspicions. Double refraction in isotropic media (which we will call nonlinear birefringence) and the Raman effect, in which a scattered wave whose frequency was the sum or difference of the frequencies of an applied field and a natural medium vibration, could not be explained on a linear basis and were indicators that the field intensity was perhaps important after all, but, for the most part, optics seemed to be a linear science. Where were the "far from linear" behaviors so richly manifested in other fields, the magnificent ferocity of a twenty-foot breaking water wave, the shock-wave boom of a supersonic jet, the majestic and thunderous cumulus cloud, the sudden implosion of a compressed shell, the surge of current in vacuum tubes at a critical applied voltage? They were there, all right, but were hidden because of the relatively low intensities that occurred naturally (e.g., sunlight, 600 volts per meter) or could be attained in the laboratory. Each was so much less than the binding fields of the hydrogen atom (10^{11} volts per meter) that it looked like the nonlinear tiger in optics would be forever contained.

The discovery of the laser in 1960 (an acronym for light amplification by stimulated emission of radiation), which was the natural lightwave analog of the maser (substitute "microwave" for "light") developed in the early 1950s, changed all that. Now available was a source of highly coherent radiation that could be concentrated and focused to give extremely high local intensities (the latest laser pulses have peak intensities of up to 10^{18} watts per square centimeter!). The nonlinear tiger was released from its cage; a rich stream of fundamental new phenomena, plus several new manifestations of phenomena familiar from other fields, soon followed, and that stream continues to flow, becoming richer by the day. The relatively young subject of nonlinear optics, the study of how high-intensity light interacts with and propagates through matter, is so scientifically fertile and technologically promising that it is destined to be one of the most important areas of science for the next quarter century.

The field of nonlinear optics is partially driven by anticipation of enormous technological dividends. Already the use of lasers in modern technology is commonplace, ranging in application from high-density data storage on optical disks to greatly improved surgical techniques in ophthalmology, neurosurgery, and dermatology. However, for future uses, more sophisticated understanding will be required. Lasers are not the stable output intensity devices suggested by the simple models. In reality, they are highly complicated dynamical systems, which can display the full range of dynamical behavior, from the staid to the exotic, from fixed-point attractors to chaotic attractors, and the nature of this complicated behavior needs to be categorized and understood. The coupling of lasers is also important, in particular in semiconductor devices where hundreds of miniature lasers can be fabricated on a single substrate, providing in principle the capability of coherent high power output from a compact solid-state laser. But how will these arrays work in practice? Can they indeed be coupled in such as way as to give a phase-locked and coherent output? Nobody really knows. Preliminary models suggest that in many parameter ranges, the array will not behave as a single coherent unit but will display a rich mosaic of spatiotemporal patterns.

Nonlinear optics is also likely to revolutionize future telecommunications and computer technologies. The relatively long interaction lengths and small cross-sections available in waveguide and fiber materials means that low-energy optical pulses can achieve sufficiently high peak intensities to compensate for the intrinsically weak nonlinearities in many transparent optical materials. Today, using linear propagation methods, information races across continents and oceans on optical fibers as thin as a human hair at rates of gigabits (10^9 bits) per second. There is every indication that within the decade, the linear technology will be replaced by a nonlinear one in which trains of light pulses are transmitted as solitons. The enormous bandwidth for data transmission afforded by optical fibers allows much scope for original ideas on all-optical controlled multiplexing (integrating bits of information carried on different channels or wavelengths) of data bits represented as soliton envelope pulses containing light at different wavelengths in a single fiber. Directional couplers, which exploit the overlap of evanescent tails of transversely guided field profiles, can switch soliton pulses from one fiber to its neighbor precisely because of the soliton's coherence. The semiconductor laser arrays mentioned earlier may well serve as the pump sources for the pulses used in optical fibers. They are also possible candidates for the power sources in all-optical signal processing and computing devices. The ultrafast, massively parallel (i.e., laser beams do not interact in free space), and global connectivity features make optical architecture an attractive alternative for computation. This architecture will likely exploit bistable behavior in optical feedback systems, such as ring and Fabry–Perot cavities, as the basic logic element. Lasers, communications, computing, image storage, beam cleanup ... the list of opportunities for useful applications of nonlinearity in optics goes on and on.

It is an ideal subject for the theoretician interested in nonlinear behavior and model building who is particularly well positioned to make major contributions to the development of the subject. First, it is incredibly diverse in that it displays the full spectrum of behavior associated with nonlinear equations, three- and four-wave resonant interactions, self-focusing, the development of singularities and weak solutions, solitons, pattern formation, phase locking, strange attractors, homoclinic tangles, the full range of bifurcation scenarios, turbulence—all familiar to the theoretician in a variety of contexts. Second, modeling or the art of the judicious approximation, the skill to recognize and then transfer ideas that run parallel in other fields, and the ability to see how the parts fit into a unified whole are key ingredients for success. Third, several new concepts of nonlinear science, including the *soliton* and the *strange* *attractor,* representing ideals at the opposite ends of the spectrum of dynamical behavior, are often encountered and require some depth of mathematical knowledge to understand. The soliton was discovered, with the aid of the computer experiment, by the mathematicians Kruskal and Zabusky, and we shall see in this book how this robust object is likely to play an ever-increasing role in propagation in fibers, surface waves in nonlinear dielectrics, switching in waveguides, optical bistability, and propagation through inhomogeneously broadened resonant media. The strange attractor was discovered by Lorenz, an atmospheric scientist with a distinctly mathematical bent, and developed as a fundamental concept in the theory of dynamics of dissipative systems by Ruelle and Takens. This idea, together with a revolution in our understanding about the nature of finite and low-dimensional dynamical systems, has had and will continue to have a broad impact, particularly in optical feedback devices such as lasers. Last but not least in the arsenal of tools the theoretician brings to bear are the techniques of modern computer simulation. Ideas and theories can be tested throughout whole parameter ranges, and quantitative support can be given to complement the qualitative understanding obtained through the use of general arguments and simple models. These attributes and tools, when combined with physical intuition and the keen realist's eye of the experimentalist, are and will continue to be the engines that drive the subject's development.

What do we mean when we use the word nonlinear or talk about nonlinear science (optics, mechanics, physics, waves, etc.) as a subject? The literal meaning "pertaining to things not linear" is not really satisfactory because it defines a subject by what it is not; and on the surface it has about the same degree of vagueness as a description of all American animals as "nonelephant." It would include all relations between quantities the graphs of which are not straight lines, and this interpretation covers many natural phenomena. Nevertheless, although it is difficult to come up with a precise definition, one can clearly indicate what the word and subject connote. In the section that follows, we discuss briefly the ingredients that make nonlinear systems so different. Generally one finds that the solutions or output data of nonlinear systems display behaviors that depend very sensitively on input data and parameters, and it is therefore very difficult not only to obtain expressions for the former in terms of the latter but even to gain any understanding using only analytical methods.

How is it, then, that nonlinear optics is so accessible to theoretical analysis, especially when compared to other branches of nonlinear physics? The key reasons are that (1) at currently available light intensities, the nonlinear coupling coefficients are small, and (2) the power spectrum of the electromagnetic field is concentrated in the neighborhoods of discrete frequencies. These properties allow one to remove all fast space (10^{-6} m) and time scales (10^{15} s) from the equations using standard perturbation techniques, and this leads to considerable simplification. To a first approximation, light and matter can be considered as a system of uncoupled oscillators; light consists of wavetrains $\exp i(\vec{k} \cdot \vec{x} - \omega t)$ while the oscillators of matter are electronic transitions, molecular vibrations and rotations, and acoustic waves. Therefore, to a first approximation, the variables of light and matter obey linear equations and the nonlinear terms are an order of magnitude smaller. This does not mean that they have negligible long-time and long-distance effects. Weak coupling does mean, however, that only certain identifiable subsets of all possible linear and nonlinear interactions between the oscillators are important, namely the small number of sets that satisfy special resonance conditions. Because of this, the fields can be accurately approximated over long times and distances by a *finite* combination of oscillator modes, namely those modes that take part in resonant interactions. The loss of energy to the many other degrees of freedom can be accounted for by attenuation terms (called homogeneous broadening) that are linear in the matter variables. Further, since the spectra of the electromagnetic and matter fields are localized in wavenumber-frequency space, the fields can be represented as finite sums of discrete wavepackets, $A(x, y, z, t) \exp i(lx + my + kz - \omega t) + (*)$, where $*$ is the complex conjugate, the wavevector components (l, m, k) are related to the frequency ω through the dispersion relation, and the amplitude $A(x, y, z, t)$ is a slowly varying function of space and time, that is, $\partial^2 A/\partial t^2 \ll \omega \partial A/\partial t \ll \omega^2 A$, $\partial^2 A/\partial z^2 \ll k \partial A/\partial z \ll k^2 A$. This means that the envelope A of the wavepacket obeys an equation containing only low powers of the derivatives $\partial/\partial t$, $\partial/\partial x$, $\partial/\partial y$, $\partial/\partial z$. Typically, the basic time and distance units for light waves in the visible range, $2\pi\omega^{-1}$ and $2\pi k^{-1}$, are of the order of a femtosecond (fs, or 10^{-15} s) and a micrometer (μm, or 10^{-6} m) respectively. The times over which the amplitudes vary lie in the range between nanoseconds (ns, or 10^{-9} s) and picoseconds (ps, or 10^{-12} s), comparable to the inverse of the magnitude of the coupling coefficient or the width of the wavepacket.

A principal goal of theory is to write down envelope equations for these amplitudes. These equations are nonlinear, but they often fall into categories of nonlinear equations about which much is known. Whereas in Chapters 2, 3, and 6 we introduce the standard perturbation procedures for deriving them from the governing Maxwell's equations, one can readily deduce what the inviscid or frictionless form of the envelope equations must be by applying simple symmetry arguments. For example, suppose one is following the evolution of a single wavepacket

$$\vec{E}(\vec{x},t) = \hat{e}(\, A(\vec{x},t)\exp i(\vec{k}\cdot\vec{x}-\omega t) + *), \quad \vec{x} = (x,y,z), \quad \vec{k} = (l,m,k), \tag{1.1}$$

in a nonlinear dielectric with a centrosymmetric crystal structure (the crystal has reflection symmetry about the origin) so that if $\vec{E}(\vec{x},t)$ is a solution so is $-\vec{E}(\vec{x},t)$. Therefore, if A satisfies the envelope equation, so does $-A$. The equation for $\vec{E}(\vec{x},t)$, derived directly from Maxwell's equations, has the additional properties of space and time reversibility and translation, meaning that if $\vec{E}(\vec{x},t)$ is a solution, so are $\vec{E}(-\vec{x},-t)$ and $\vec{E}(\vec{x}+\vec{x}_0, t+t_0)$ for arbitrary $\vec{x}_0, t_0$. The last two properties mean that if $A(\vec{x},t)$ is a solution of the equation which the envelope satisfies, so are $A^*(-\vec{x},-t)$ and $A(\vec{x},t)\exp i\phi_0$ for arbitrary constant ϕ_0. In addition, the fact that we are dealing with *weak* nonlinear coupling of *long* wavepackets means that the equation for the amplitude A is a multinomial expansion in powers of A and the gradient $\nabla(\partial/\partial x, \partial/\partial y, \partial/\partial z)$ and time derivative $\partial/\partial t$ operators. To leading order, the only nontrivial candidate satisfying all these properties must be a real, linear combination of $\partial A/\partial t$, ∇A, i times all possible second derivatives, iA, and $i|A|^2A$, precisely the terms that compose the universal and ubiquitous nonlinear Schrödinger (NLS) equation. If A is constrained to depend on one direction only, as in a light fiber, its equation will be

$$\frac{\partial A}{\partial z} + k'\frac{\partial A}{\partial t} + \frac{i}{2}k''\frac{\partial^2 A}{\partial t^2} - i\frac{\delta n(|A|^2)}{n}kA = 0, \tag{1.2}$$

with the refractive index correction $\delta n/n$ given by the first two terms $a + b|A|^2$ in a Taylor expansion in the field intensity and where the coefficients k, k', and k'' all have a very natural interpretation. In Chapter 2, we show that the operator that gives rise to the multinomial is simply the *dispersion relation*, namely the equation that relates the wavevector $\vec{k}$, frequency ω, and intensity $|A|^2$ of a nonlinear wavetrain. Equation (1.2) is the one-dimensional NLS equation (the dimension of the NLS equation is defined by the number of variables appearing as second derivatives), which has very special mathematical properties reflected in a wonderful class of solutions called *solitons*. The soliton is to nonlinear science what the Fourier mode is to linear science, namely a fundamental "normal" mode of propagation of a nonlinear system, and we have much to say about it in this book.

Is it a fluke that many of the equations of mathematical physics like the NLS equation or the Korteweg–de Vries equation or the sine–Gordon equation, which are derived by standard perturbation analyses as the universal asymptotic description of a wide variety of physical systems, are integrable or close to being integrable? We don't know the answer to this, but the following comment may be relevant. A key observation is that if one starts with an exactly integrable system, then the asymptotic analysis leading to the equation that describes the long-time behavior of the envelopes of special types of solutions does not destroy this integrable character; rather, the integrability is preserved. Therefore if among the set of all equations that reduce to the same asymptotic description there is one equation that is integrable, then the asymptotic equation is integrable. Therefore whereas integrability is rare in general, the process of reduction to universal, asymptotic equations for wave envelopes increases the probability that the resulting equation has special properties. The reduction process introduces new symmetries and new constraints (conservation laws) and does not destroy existing ones.

For the most part, therefore, the theorist can gain access to nonlinear optics by decomposing the relevant field variables into a finite basis of weakly interacting wavepackets, and the evolution of the field is obtained by following the *fully nonlinear* evolution of the wavepacket envelopes, which equations, because of symmetries and their universal nature, tend to have rather special properties. Being able to get off the ground with a finite linear basis is a luxury denied to most other areas of continuum physics, like fluid mechanics for example, in which nonlinearity is strong and for which in most circumstances it is impossible to neglect any higher-order interactions and approximate the fields uniformly in time and space by a finite number of modes. In a sense we can think of the NLS equation as being the optical analog of the Euler equations, the high-Reynolds-number, inviscid limit of the Navier–Stokes equations, with the one (two) dimensional NLS equation corresponding to the two (three) dimensional Euler equations. Much more information is known about the NLS equation in both cases. Of course, when light intensities are so high they raise the temperature of matter to a point where it behaves like a plasma, then the weak coupling theory is no longer valid. Up to that point, however, there remains much to be explored and discovered in contexts where the weak coupling approximation is applicable.

For systems like lasers, which are confined by finite geometries, the spectrum is discrete, spatial shapes are determined, and the spatial gradient terms disappear. Again, however, because of the constraints of resonances, the dynamics is described by a relatively low-order set of ordinary differential equations of dissipative type for the oscillator amplitudes. The dissipation arises from two sources. The electric field loses energy mainly because of imperfect mirrors. Matter loses energy because of collisions and the slow irreversible decay of the number of excited atoms to levels that are not included in the approximation. These effects, called homogeneous broadening, are modeled by the inclusion of linear damping terms. As we have mentioned, the behavior of the solutions of these coupled sets of ordinary differential equations can be very complicated. While there is no general theory for determining the nature of the asymptotic states (whether fixed points, limit cycles, or strange attractors) for such sets of equations, there has nevertheless been built up much qualitative experience about their behavior. This has been greatly facilitated by the new generation of computer workstations with their high performance processing and graphical capabilities, which provide a very powerful analytical research tool. In Chapter 6, we describe some of the sophisticated software available on most modern computer workstations. In addition to the standard commercial scientific subroutine packages such as IMSL or NAG, and symbolic programming languages like MACSYMA or REDUCE for algebraic manipulation of complex expressions, one can gain access to bifurcation packages (AUTO, PITCON, etc.) and a host of very powerful UNIX-based public domain software. The student can, with little computational effort, realize the marvelous and deep mathematical insights of Poincaré regarding the geometry of phase space in real time on a high-resolution graphics screen. For example, numerical integrators for the systems of ordinary differential equations (o.d.e.'s) modeling standard laser systems can be found in the standard IMSL or NAG packages available on most workstations.

We begin in the next section by briefly describing behavior unique to nonlinear systems. We then discuss dielectrics and emphasize that the principal medium variable, the susceptibility or refractive index, which tells us how the polarization field depends on the applied electric field, is almost impossible to calculate in general. In a very real sense, therefore, we start from a constitutive relation that is at best an approximation. Consequently, it is clear that a top priority of the field is to develop a better understanding of how to calculate the susceptibility and how to design materials with susceptibilities having advantageous properties. We then briefly discuss two areas of great importance that are not treated in this edition, namely mode locking and pulse compression and the interaction of light with semiconductors, each of which has enormous practical potential and intellectual challenges. We felt that to omit any mention of them would signal that they were not important. To disabuse the reader of that conclusion, we decided to mention them up front.

The Nature of Nonlinearity

We look at the question of defining a nonlinear system two ways, first by the mathematical character of the laws (equations) that describe its behavior