NONLINEAR OPTICS

LECTURE NOTE AND REPRINT VOLUME

N. BLOEMBERGEN

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EDITOR'S FOREWORD

The problem of communicating in a coherent fashion the recent developments in the most exciting and active fields of physics seems particularly pressing today. The enormous growth in the number of physicists has tended to make the familiar channels of communication considerably less effective. It has become increasingly difficult for experts in a given field to keep up with the current literature; the novice can only be confused. What is needed is both a consistent account of a field and the presentation of a definite "point of view" concerning it. Formal monographs cannot meet such a need in a rapidly developing field, and, perhaps more important, the review article seems to have fallen into disfavor. Indeed, it would seem that the people most actively engaged in developing a given field are the people least likely to write at length about it.

"Frontiers in Physics" has been conceived in an effort to improve the situation in several ways. First, to take advantage of the fact that the leading physicists today frequently give a series of lectures, a graduate seminar, or a graduate course in their special fields of interest. Such lectures serve to summarize the present status of a rapidly developing field and may well constitute the only coherent account available at the time. Often, notes on lectures exist (prepared by the lecturer himself, by graduate students, or by postdoctoral fellows) and have been distributed in mimeographed form on a limited basis. One of the principal purposes of the "Frontiers in Physics" series is to make such notes available to a wider audience of physicists.

It should be emphasized that lecture notes are necessarily rough and informal, both in style and content, and those in the series will prove no exception. This is as it should be. The point of the series is to offer new, rapid, more informal, and, it is hoped, more effective ways for physicists to teach one another. The point is lost if only elegant notes qualify.

A second way to improve communication in very active fields of physics is by the publication of collections of reprints of recent articles. Such collections are themselves useful to people working in the field. The value of the reprints would, however, seem much enhanced if the collection would be accompanied by an introduction of moderate length, which would serve to tie the collection together and, necessarily, constitute a brief survey of the present status of the field. Again, it is appropriate that such an introduction be informal, in keeping with the active character of the field.

A third possibility for the series might be called an informal monograph, to connote the fact that it represents an intermediate step between lecture notes and formal monographs. It would offer the author an opportunity to present his views of a field that has developed to the point at which a summation might prove extraordinarily fruitful, but for which a formal monograph might not be feasible or desirable.

Fourth, there are the contemporary classics—papers or lectures which constitute a particularly valuable approach to the teaching and learning of physics today. Here one thinks of fields that lie at the heart of much of present-day research, but whose essentials are by now well understood, such as quantum electrodynamics or magnetic resonance. In such fields some of the best pedagogical material is not readily available, either because it consists of papers long out of print or lectures that have never been published.

"Frontiers in Physics" is designed to be flexible in editorial format. Authors are encouraged to use as many of the foregoing approaches as seem desirable for the project at hand. The publishing format for the series is in keeping with its intentions. Photo-offset printing is used throughout, and the books are paperbound, in order to speed publication and reduce costs. It is hoped that the books will thereby be within the financial reach of graduate students in this country and abroad.

Finally, suggestions from interested readers as to format, contributors, and contributions will be most welcome.

DAVID PINES

Urbana, Illinois
August 1964

PREFACE

This monograph is based on lectures, prepared for a course on quantum electronics at Harvard University in the spring of 1963 and for the summer school in Les Houches in 1964. The field of nonlinear optics is quite young. It deals with phenomena that occur at very high light intensities obtainable in laser beams. It represents one of the most interesting fields of research made possible by the development of powerful lasers.

It is perhaps foolhardy to write a monograph about nonlinear optics at this time, when new results are still announced at a high rate in scientific journals. It could be argued that such a monograph would at best contribute to its own rapid obsolescence. Nevertheless, it is hoped that it may have some more lasting value. The general principles of Maxwell's electromagnetic theory and of quantum mechanics are well established. Their domain of application is extended to include higher order interactions between light and matter in terms of nonlinear susceptibilities.

The nonlinear response of circuit elements at audio-radio and microwave frequencies is well known to the electrical engineer. In this monograph the analogous phenomena at optical frequencies are discussed. The concepts of harmonic generation, parametric amplification, modulation and rectification all have their counterparts in the visible region of the electromagnetic spectrum. The material is organized so that a pure classical description can be followed by those who have a knowledge of electromagnetic theory but are not familiar with quantum mechanics. They may skip Chapter 2 in which the quantum theory of linear and nonlinear susceptibilities is treated. This volume is intended for all who have an active interest in the field of quantum electronics, whether they are physicists interested in nonlinear electromagnetic properties of matter, electrical engineers interested in communications or high power applications at visible frequencies, or optical scientists interested in the behavior of light rays at very high intensities.

Since the field of nonlinear optics is still in a stage of rapid expansion, no effort has been made to give a complete bibliography nor to achieve a complete coverage of all experimental data. The fundamental theoretical ideas and the basic experimental results are emphasized.

The author is indebted to Dr. J. Ducuing, Dr. Y. R. Shen, and Dr. D. Forster who have carefully read the manuscript and suggested many corrections. Any errors that remain are entirely the responsibility of the author. The permission of the editors of *The Physical Review* and the respective coauthors to reproduce the three appendices is gratefully acknowledged. The author is indebted to Drs. P. S. Pershan and R. W. Terhune for making available some material before publication. The author wishes to express his thanks to Elizabeth Dixon who typed the entire manuscript on a tight schedule.

This monograph is dedicated to Deli Bloembergen, whose encouragement and understanding were a decisive factor in its timely completion.

N. Bloembergen

Cambridge, Massachusetts
July 1964

PREFACE TO THE THIRD PRINTING

The field of nonlinear optics has come of age. At this time the published literature in this field is at least two orders of magnitude larger than in 1964, when these lecture notes were produced. Obviously, a better and more comprehensive textbook could now be written. Lacking time and effort to accomplish that needed task, I am gratified that these original notes still provide a general framework, suitable for the description of many new developments in nonlinear optics. This reprint should satisfy an apparent demand which does not originate purely in historical curiosity. The addition of an Epilogue points out some of the shortcomings of these notes, but also shows how they provide a useful introduction to modern developments. The addition of these comments plus a selected bibliography of recent books and review papers should be helpful to the student who wishes to reach – or the more advanced worker, who is already engaged in – the current frontiers of research and engineering developments in nonlinear optics.

N. Bloembergen

Cambridge, Massachusetts
Fall, 1976

EXPLANATORY NOTES FOR THE THIRD PRINTING

A bibliography covering more recent developments not included in the reprint of the main text, originally written in 1964, is added in the Epilogue of the present printing. The present book can still be used as a self-contained discussion and introduction to those fundamental principles of nonlinear optics which are describable in terms of nonlinear susceptibilities. Although no attempt has been made to update the material nor to correct all minor misprints and errors, the following notes will eliminate some difficulties which past readers have experienced in several passages.

Page 3, Eq. (1-3) and following. The definition of the amplitude adopted in this book has not survived. A definition with a factor 1/2 inserted on the right hand side of Eq. (1-3) is now in common usage. A change in definition necessitates, of course, corrections by one or more factors of two in many locations before the results here can be compared with those in other publications.

Page 5, Eq. (1-8). The complex conjugate expression should be added to the right hand side of Eq. (1-8), which should read:

$$\frac{2N_0 e^3 \, |E|^2}{m^2 c(\omega^2 + \tau^{-2})}.$$

The two sentences following this equation should be deleted.

Page 6. A factor 2 should be added to the right hand side of Eq. (1-2). This factor is due to the two permutations of the amplitudes E_1 and E_2^* in the double product of $(E_1 + E_2^*)^2$. These and other permutation degeneracy factors are carefully discussed, for example, by S. K. Kurtz in *Quantum Electronics*, ed. H. Rabin and C. L. Tang, Vol. 1A, p. 209, Academic Press, New York, 1975.

Page 7. Eqs. (1-15) and (1-18) need an additional degeneracy factor of 6 on the right hand side.

Page 28. The paragraph of lines 4-20 from the top should be replaced by the following:

> "The correct limiting behavior for the case that either the electromagnetic frequency or the material resonant frequency becomes very small, $\omega \to 0$ or $\omega_{ng} \to 0$, respectively, requires a more careful treatment of the damping terms, as has been discussed in detail by Van Vleck and Weisskopf.[3]"

Page 30. A minus sign should be added to the right hand side of Eq. (2-34).

Page 57. "ω_{ab}" should be replaced by "ω_{ba}" on the right side of Eq. (2-98).

Page 72. In Eq. (3-26) "dE" should be replaced by "$-$dE".

Page 73. In Eq. (3-30) "f_r" should be replaced by "f_R".

Pages 121-165. The experimental results and numerical data quoted in Chapter 5 are in need of considerable revision and updating. For such information the reader is referred to the literature quoted in the Epilogue.

Page 198. A factor ϵ_R^{-1} should be added to the right hand side of the second expression (Eq. 4.5).

Page 199. In the denominator of the last term on the right hand side of Eq. (4.12), $\epsilon_S^{1/2}$ should be replaced by $\epsilon_R^{1/2}$.

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The publisher wishes to thank the American Institute of Physics for permission to reprint material from *The Physical Review.*

1

CLASSICAL INTRODUCTION

1-1 NONLINEAR SUSCEPTIBILITIES

Nonlinear properties of Maxwell's constitutive relations,

$$D = \epsilon(E)E \qquad B = \mu(H)H \tag{1-1}$$

have been known from the beginning. The dielectric constant and magnetic permeability can be functions of the field strengths. The nonlinear permeability of ferromagnetic media was of prime concern in the design of electrical machinery in the nineteenth century. This nonlinearity has its origin in domain wall motion and domain rotation. It can be a source of harmonic distortion in audio-amplifiers using inductances with ferromagnetic cores. Paramagnetic saturation described by Langevin and Brillouin may be considered as a nonlinearity at zero frequency. Magnetic and dielectric amplifiers are based on the nonlinearity of ferromagnetica and ferroelectrica at relatively low frequencies. The nonlinear electromagnetic response of a plasma has been known for a long time. It was invoked to explain the "Luxembourg" effect in ionospheric propagation of radiowaves. More recently, the nonlinear properties of plasmas and other materials have been investigated at microwave frequencies. The nonlinearity in ferromagnetic resonance, for example, has been used to generate second and higher harmonics in the microwave region of the electromagnetic spectrum.

Nonlinear properties in optical region have been strikingly demonstrated by harmonic generation of light. Franken[1] and coworkers detected ultraviolet light (λ = 3470 Å) at twice the frequency of a ruby laser beam (λ = 6940 A) when this beam traversed a quartz crystal. A schematic diagram of the experimental arrangement

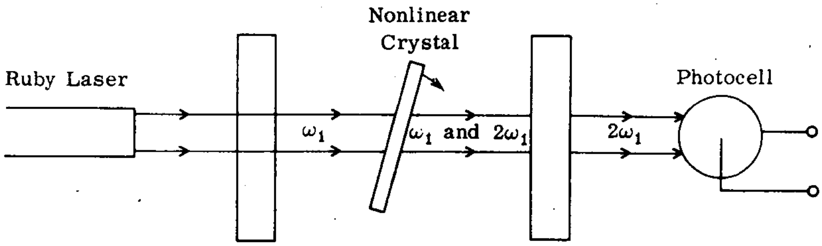

Figure 1-1. Experimental arrangement for the detection of second harmonic generation of light.

is shown in Figure 1-1. This experiment, carried out in 1961, marked the beginning of a large activity in both experimental and theoretical nonlinear optical properties. This work has been reviewed in a number of papers,[2,3,4] where the reader may find rather complete references to the literature. In this volume no attempt will be made to give a comprehensive review of all the detailed work in this field of endeavor, but emphasis will be placed on the development and discussion of the diverse phenomena from a unified point of view. The nonlinear material properties are described by expanding the polarization in a power series in the field. For the pure electric dipole case one has, for example,

$$\mathbf{P} = \chi \cdot \mathbf{E} + \chi : \mathbf{EE} + \chi \vdots \mathbf{EEE} + \cdots \tag{1-2}$$

The first term defines the usual linear susceptibility, the second term, the lowest order nonlinear susceptibility, and so on. This procedure is useful because the optical nonlinearities are small. In fact, their very small size is largely responsible for their late experimental discovery, which had to wait for the development of powerful lasers. Large electric fields with peak amplitudes of about one million volts/cm, corresponding to a flux density of about 10^9 watts/cm^2, are now available at optical frequencies from Q-switched lasers.[5] Effects are under favorable conditions already detectable with a flux density of less than one mw/cm^2.

The theory of wave propagation in nonlinear media can be developed along purely classical lines and the nonlinear optical properties can be discussed along the same lines as the linear optical properties at the turn of the last century. Maxwell, Hertz, Lorentz, and Drude, however, lacked the stimulation of experimental findings. The advent of stimulated emission of light has suddenly changed this. The behavior of light beams at high intensity is studied vigorously.

Many generalizations of the classical laws of optics to the regime of intensities, where nonlinearities are important, have already been made. At the same time the nonlinear susceptibilities themselves are of intrinsic interest in the study of the structure of matter. This situation is of course similar in the linear case. The linear index of refraction determines the paths of light rays. Conversely, knowledge of its behavior gives information about the nature of the material.

1-2 CLASSICAL ATOMIC MODELS OF NONLINEARITY

The Free Electron Gas

Consider the motion of a single electron in a plasma under the influence of a linearly polarized light wave:

$$\begin{aligned} B_y = E_x &= E \exp(ikz - i\omega t) + E^* \exp(-ikz + i\omega t) \\ &= 2 \,\mathrm{Re}\,\{E \exp(ikz - i\omega t)\} \text{ with } k = \omega c^{-1} \end{aligned} \tag{1-3}$$

Note that the real amplitude of the wave is $2|E|$, or our amplitude $|E|$ is half that of the usual definition. This new convention, introduced by Pershan,[4] leaves the linear susceptibility unchanged, but increases the lowest order nonlinear susceptibility by a factor of two. Additional factors of two appear in higher order susceptibilities. The new convention has a definite advantage in the discussion of certain symmetry properties of nonlinear susceptibilities. The limiting cases that two frequencies become equal, or one of the frequencies goes to zero, have been a source of confusion.[6] The new system largely avoids difficulties with factors two during the computation. One may revert to the old convention at the end of the calculation. The Fourier component of a physical quantity at the frequency $+\omega$ has a time dependence $\exp(-i\omega t)$, the Fourier component at $-\omega$ goes as $\exp(+i\omega t)$.

The equations of motion for a single electron in the plasma are,

$$\begin{aligned} m\ddot{x} &= eE_x - ec^{-1}\dot{z}B_y - m\dot{x}/\tau \\ m\ddot{y} &= -m\dot{y}/\tau \\ m\ddot{z} &= ec^{-1}\dot{x}B_y - m\dot{z}/\tau \end{aligned} \tag{1-4}$$

The phenomenological collision time τ describes the damping of the motion in a statistical sense and insures a steady state response independent of the initial conditions. The Lorentz force gives rise to harmonics. A steady state solution of Eqs. (1-3) and (1-4) can be found by successive approximation in the form of a Fourier series.

In the first or linear approximation one has the well-known result

$$x(\omega) = \frac{-eE\exp(ikz - i\omega t)}{m(\omega^2 + i\omega\tau^{-1})} \tag{1-5}$$

If this linear solution is substituted into the last equation of (1-4), the lowest order nonlinear approximation gives

$$z(2\omega) = \frac{-ie^2E^2\exp(2ikz - 2i\omega t)}{m^2c(4\omega + 2i\tau^{-1})(\omega^2 + i\omega\tau^{-1})} \tag{1-6}$$

The linear dipole moment $ex(\omega)$ gives rise to the well-known Thomson formula, describing the Rayleigh scattering of light by free electrons. In a similar way the dipole moment $ez(2\omega)$ radiates at the second harmonic frequency. This nonlinear scattering process may roughly be described by saying that two incident quanta are taken away from the incident beam and one quantum at twice the frequency is radiated with the intensity pattern of a dipole oriented along the direction of the incident beam.

The incoherent nonlinear scattering by individual electrons is, however, not of practical interest. The nonlinear phenomena are sufficiently weak that only the coherent radiation of a large assembly of particles is detectable. A similar distinction between coherent and incoherent scattering holds for the linear properties of a plasma. The attention should be focused on the average polarization in a small volume and the index of refraction of the plasma, rather than on the incoherent Rayleigh scattering due to fluctuations in density. In liquids and crystals the relative density fluctuations are even smaller and the main interest is in the coherent polarization of the medium.

If the average density of electrons in the plasma is N_0 per cm^3, the polarization is

$$P_x(\omega) = \chi(\omega)E_x(\omega) = N_0ex(\omega). \tag{1-7}$$

Since at optical frequencies $\omega\tau \ll 1$, Eqs. (1-5) and (1-7) give immediately the familiar result for the susceptibility of a plasma:

$$\epsilon - 1 = 4\pi\chi = -4\pi N_0e^2/m\omega^2$$

In a similar way, the nonlinear polarization at the second harmonic frequency is given by

$$P_z(2\omega) = N_0ez(2\omega).$$

There is no coherent radiation at 2ω in an infinite plasma, because this polarization is parallel to the direction of propagation. Coherent second harmonic radiation is possible at the boundary of a plasma.

There is also a small dc current in the longitudinal direction in this approximation, generated by the term $\dot{x}(\omega)B_y(-\omega) + \dot{x}(-\omega)B_y(+\omega)$. One finds the dc current density

$$J_z(o) = N_0 e\dot{z}(o) = \frac{iN_0 e^3 \omega\tau |E|^2}{m^2 c(\omega^2 + i\omega\tau^{-1})} \tag{1-8}$$

This term represents the classical analogue to momentum transfer in Compton scattering.

The procedure can of course readily be extended to higher harmonics. There is a component of polarization $P_x(3\omega)$ which will generate third-harmonic radiation.

The nonlinearities in plasmas at microwave frequencies can be quite large, especially in the presence of a dc magnetic field. Then cyclotron resonance effects can occur.[7] In actual plasmas at lower frequencies other nonlinearities, besides the Lorentz force, are important. Hydrodynamic pressure gradients and induced variations in the electron density must be considered. A rather complete discussion of all these effects has been given by Whitmer and Barrett.[8]

The Anharmonic Oscillator

A very useful model used by Drude and Lorentz[9] to calculate the linear polarization of a medium describes the electrons as harmonically bound particles. The resonant frequencies of the oscillators were taken to correspond to the observed atomic spectral lines. Actually the valence electrons are bound by the Coulomb field of the ion cores. For very large deviations from equilibrium the anharmonicity of the electron oscillators must be taken into account. Such a model had already been used by Rayleigh to explain nonlinearities in acoustic resonators.[10] Consider therefore the motion of a one-dimensional anharmonic oscillator with damping, driven by an electric field with Fourier components at the frequencies $\pm\omega_1$ and $\pm\omega_2$.

$$\ddot{x} + \Gamma\dot{x} + \omega_0^2 x + vx^2 = (2e/m)\,\mathrm{Re}\{E_1 \exp(ik_1 z - i\omega_1 t) + E_2 \exp(ik_2 z - i\omega_2 t)\} \tag{1-9}$$

The linear approximation gives immediately the well-known result

$$x(\omega_1) = \frac{e}{m(-\omega_1^2 + \omega_0^2 - i\omega_1\Gamma)} E_1 \exp(ik_1 z - i\omega_1 t) \tag{1-10}$$