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Nonlinear Optics in Solids

O. Keller (Ed.)

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Preface

In recent years one has witnessed in physics a substantial increase in interest in carrying out fundamental studies in the nonlinear optics of condensed matter. At the Danish universities, this increase has been especially pronounced at the Institute of Physics at the University of Aalborg, where the main activities are centered around fundamental research within the domains of nonlinear quantum optics, nonlinear optics of metals and superconductors, and nonlinear surface optics. In recognition of this it was decided to arrange the first international summer school on nonlinear optics in Denmark at the Institute of Physics at the University of Aalborg.

This book is based on the lectures and contributed papers presented at this international summer school, which was held in the period 31 July–4 August 1989. About 60 experienced and younger scientists from 12 different countries participated. Twenty-eight lectures were given by 14 distinguished scientists from the United States, Italy, France, Germany, Scotland, England, and Denmark. In addition to the lectures given by the invited speakers, 11 contributed papers were presented. The programme of the summer school emphasized a treatment of basic physical properties of the nonlinear interaction of light and condensed matter and both theoretical and experimental aspects were covered. Furthermore, general principles as well as topics of current interest in the research literature were discussed. Among the topics included in the summer school were the following: optical bistability, principles and applications; enhanced second-harmonic generation from metallic surfaces; nonlinear effects in guiding structures, thin films and single interfaces; nonlinear optics of semiconductor quantum wells; field quantization in quantum optics and production of nonclassical light via nonlinear processes; optical phase conjugation, photorefractive effects; quantum electrodynamics in cavities, nonlinear optical effects in organic materials; light pressure on single particles, unified treatment of parametric phenomena and nonlinear ponderomotive effects in condensed matter; nonlinear optics of centrosymmetric metals and Cooper-paired superconductors; transient nonlinear optics of semiconductors; sum- and difference-frequency generation, parametric processes and harmonic generation; second-harmonic generation and nonlinear pulse generation in optical fibres; inelastic light scattering from solid surfaces, in particular Brillouin scattering; three- and four-wave mixing in disordered nonlinear media and enhanced backscattering; and critical phenomena in cooperative Raman scattering.

The invited speakers covered the following subjects: I. Abram: The nonlinear optics of semiconductor quantum wells; A.D. Boardman: Nonlinear effects in guiding structures: thin films and interfaces; J.-L. Coutaz: Surface enhanced second-harmonic generation from metals; T.J. Hall: Phase conjugation in solids; J. Hvam: Transient nonlinear optics in semiconductors; O. Keller: Nonlinear optics of centrosymmetric superconductors; P.L. Knight: Field quantization in quantum optics and production of nonclassical light; P. Meystre: Cavity quantum electrodynamics; P. Mulser: The role of radiation pressure in nonlinear optics; F. Nizzoli: Light scattering from solid surfaces; P.N. Prasad: Nonlinear optical effects in organic materials; T. Skettrup: Sum- and difference-frequency generation, harmonic generation and parametric processes; D.A. Weinberger: Second-harmonic generation and nonlinear pulse propagation in optical fibres; B.S. Wherrett: Optical bistability and digital optical computing.

In the initial phase of the planning of the summer school, the Institute of Physics cooperated with the Danish Optical Society. The school was sponsored by several organizations. The basic support came from the Danish Research Academy, which was established a few years ago in order to stimulate the education of young scientists in Denmark and to strengthen their contact to the international scientific community. Specific financial support was provided by Det Obelske Familiefond, Sparekassen Nordjylland, and the University of Aalborg.

Finally, I wish to express my thanks to the invited speakers, all the other participants, and coworkers at the Institute of Physics for their outstanding contributions in making the summer school stimulating, instructive, suggestive, and pleasant.

Aalborg, March 1990 *Ole Keller*

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The Exotic World of Nonlinear Optics

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It is a privilege for a physicist to work in a field which has its roots far back in the past and which has a bright future, and it is exciting if the field embraces esoteric aspects of fundamental physics as well as themes of interest to applied science. Nonlinear optics in solids dates back to the beginning of the nineteenth century when Brewster discovered the photoelastic effect in gels [1]. At the end of the century, Kerr discovered the quadratic electro-optic effect in glasses and liquids [2], and Röntgen [3] and Kundt [4] the linear electro-optic effect in quartz and tourmaline crystals. The linear electro-optic effect is often named the Pöckels effect in recognition of the important contributions made by Pöckels [5]. The above-mentioned phenomena belong to the domain of nonlinear optics since it is necessary to multiply two electric fields together to obtain the effects. The one is the high-frequency field of light, the other is the internal low (possibly dc)-frequency field associated with the deformation of a solid subjected to a mechanical stress (photoelasticity) or stemming from an externally applied electric field. In the wake of the invention of the laser thirty years ago, nonlinear optics has developed into an important and independent field of optics. By means of lasers it is possible to produce light fields of a high intensity and with a substantial degree of coherence. In turn, the use of lasers has enabled us to mix high-frequency optical fields in such manners that myriads of exotic nonlinear optical effects can be observed. Since the development of lasers possessing new outstanding properties will progress steadily in the coming years and since our understanding of the dynamics of nonlinear systems from a physical and mathematical point of view is sparse, the future of nonlinear optics will be filled with surprises and mountain tops of undreamed heights will have to be climbed. Since it is impossible in a single book to cover a substantial fraction of the subjects being studied nowadays within the domain of nonlinear optics in solids, it has been tried to bring the readers a flavour of the depth and diversity of the field by selecting a number of subjects of current interest in the research literature.

From a unified point of view, the electromagnetic interaction of light and atomic particles can be described on the basis of the wave equation for the four-vector vector potential operator $\hat{\mathcal{A}}$, i.e.,

$$\Box^2 \hat{\mathcal{A}} = -\mu_0(\hat{\mathcal{J}}^L + \hat{\mathcal{J}}^{NL}) \; , \tag{1}$$

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where $\hat{\mathcal{J}}^L$ and $\hat{\mathcal{J}}^{NL}$ are the four-vector current density operators describing the linear (L) and nonlinear (NL) interactions, respectively. The four-vectors are given by $\mathcal{A}_4 = (\vec{A}, i\Phi/c_0)$ and $\mathcal{J} = (\vec{J}, ic_0\rho)$, where $\vec{A}$ and Φ are the conventional vector and scalar potentials, and $\vec{J}$ and ρ the current and charge densities. The four-dimensional nabla operator is $\Box = (\vec{\nabla}, \frac{1}{ic_0}\frac{\partial}{\partial t})$. Operators are denoted by a caret, $\hat{}$. It has turned out in recent years that certain phenomena in the nonlinear optics of solids can be described only if the free electromagnetic field is quantized. The space-time dependence of the quantized field can be studied via the wave equation

$$\Box^2 \hat{\mathcal{A}} = 0 \tag{2}$$

for the field operator $\hat{\mathcal{A}}$. The field quantization leads to the introduction of the photon concept and the vacuum field state.

Among the physical phenomena requiring the field quantization to be understood is the well-known spontaneous emission resulting from the interaction between an atom and an electromagnetic field in the vacuum state. In a new subfield of physics named cavity quantum electrodynamics (cavity QED) the linear and nonlinear interactions between a single (or a few) atom(s) and a single (or a few) photon(s) in a resonator are studied. In the article by P. Meystre such novel effects as inhibited or enhanced spontaneous emission, micromaser action and quantum superposition in separated cavities are discussed. Since the spontaneous emission is triggered mainly by the zero-point fluctuations in the field which have frequencies in the vicinity of the atomic transition involved, for wavelengths below the cavity cutoff a drastic increase in the lifetime of the atom can be observed. As the lifetime is limited by the indirect coupling of the atom and the field via the wall it is of importance to consider the interaction of the cavity field with the many-body system of the wall.

In recent years the interaction between light and condensed matter surfaces has been extensively investigated, and in the present book several examples from the domain of nonlinear surface optics are presented. The approach adopted is semiclassical in the sense that the radiation field is considered to be a c-number. Much effort has been devoted to studies of second-order nonlinear effects in centrosymmetric media due to the fact that the dipole radiation is bulk forbidden. Since the centrosymmetry is broken at the outermost atomic layers the sum- and difference-frequency generation processes are surface sensitive. In the paper by myself, the nonlinear response function describing second-harmonic generation in centrosymmetric media is discussed within the framework of a nonlocal response theory. In a nonlocal formalism the light-induced nonlinear current density and the fundamental electric field, $\vec{E}$, are related via

$$\vec{J}^{NL}(\vec{r}) = \int_{-\infty}^{\infty} \overleftrightarrow{\Sigma}(\vec{r}, \vec{r}\,', \vec{r}\,'') : \vec{E}(\vec{r}\,'')\vec{E}(\vec{r}\,')d^3r''d^3r' \ , \tag{3}$$

where $\overleftrightarrow{\Sigma}(\vec{r}, \vec{r}\,', \vec{r}\,'')$ is a generalized nonlinear response function depending on the three space coordinates $\vec{r}, \vec{r}'$, and $\vec{r}''$. The nonlocal approach allows one to consider the competition between local surface effects and nonlocal bulk effects. As an example I apply the response formalism to a study of second-harmonic generation and optical rectification in a Cooper-paired superconductor. The even-order nonlinear responses of the centrosymmetric metals are so weak that one would like to invent schemes that enhance the nonlinear signal. In the article by J.L. Coutaz, a review of methods for surface enhanced second-harmonic generation from bare metallic surfaces is given. The experimental data presented are discussed on the basis of the free-electron model and the nonlinear optical surface response is treated within the framework of the two-parameter Rudnick and Stern model. In particular, enhancements obtained in attenuated total reflection configurations, via diffraction gratings and by means of surface roughness are discussed. Also the enhanced second-harmonic generation from metal islands is discussed.

Surfaces and interfaces also play a significant role for Brillouin and Raman scattering from opaque media. With main emphasis on Brillouin scattering from acoustic phonons and Raman scattering from surface phonon polaritons, F. Nizzoli reviews the field. In a section on experimental techniques for surface Brillouin scattering the single-pass and multi-pass Fabry-Perot interferometers and the tandem interferometer are discussed. The discrete and the continuous spectra of surface acoustic modes are treated, and also the vibrational spectra of unsupported and supported films are considered. Besides the elasto-optic effect, the surface rippling caused by the phonons contributes to the inelastic light scattering and sometimes even the interference between these two mechanisms. Finally, F. Nizzoli treats the theory and experiments related to Brillouin scattering from supported films. A better understanding of the nonlinear properties of optical fibre waveguides is of extreme importance for optical communication purposes. In the paper by D.A. Weinberger an interesting theme is treated, namely, that stemming from the recent observation that second-harmonic generation with a conversion efficiency approaching 5% can be obtained in fibres made of a centrosymmetric material like fused silica. Progress obtained to date in the understanding of this intriguing phenomenon is reviewed. Thus, electric quadrupole and magnetic dipole effects, the mixing model for self-organization of so-called $\chi^{(2)}$ gratings, the growth and erasure related to $\chi^{(2)}$ grating dynamics, and the evidence for defect structures are discussed.

A unified treatment of parametric phenomena and nonlinear ponderomotive effects in condensed matter is given in the interesting article by P. Mulser, who holds the view that the nonlinear optical phenomena can be considered as produced by the radiation pressure. This point of view seems most refreshing and it occurs to me that its consequences in the context of optical rectification in superconductors should be examined due to the fact that the light-induced dc-recoil of the Cooper-paired electrons leads to a frictionless dc-current. In 1861, Maxwell argued that a light beam of flux density I, when impinging normally from vacuum upon a surface of reflectivity R, exerts a light pressure

$$P_L = (1 + R)\frac{I}{c_0} \tag{4}$$

on the material. Taking this result as a starting point, P. Mulser presents a number of interesting historical remarks on the subject. Following the historical introduction the ponderomotive forces on single particles and in dense matter are discussed. Among the light pressure effects phenomena such as nonresonant effects, stimulated decay processes, wave pressure in moving media, and stimulated Brillouin and Stokes Raman scattering are treated. Finally, a twenty-year-old paradox is discussed.

Semiconductors play a prominent role in the nonlinear optics of solids. In the paper by I. Abram, the nonlinear optics of the new and interesting quantum wells are discussed, e.g. the observation of different types of optical nonlinearities, the physical origin of these nonlinearities and some device possibilities for optical signal processing. Among many important subjects, the optical response in the transparent regime, the resonant optical response, coherent transients, dynamical nonlinearities, and the optical Stark effect are discussed. A detailed treatment of excitonic coherent transients and of dense electron-hole populations is presented. Among possible devices the monolithic optical bistable etalon is considered. In the article by J.M. Hvam, it is described how transient optical nonlinearities are conveniently studied by time resolved excite-and-probe experiments e.g. degenerate four-wave mixing and transient laser induced grating experiments. Special emphasis is devoted to a discussion of the strong excitonic resonance enhancements of the nonlinear susceptibility observed at low temperatures in for instance CdSe. In recent years optical phase conjugation has received special attention from the scientific community. In the paper by T.J. Hall and A.K. Powell the basic principles behind phase conjugation are reviewed and the grating picture is used to demonstrate the analogy with real-time holography. The photorefractive effect is described as an example of an effect suitable for real-time holography. Interesting effects such as self-pumped phase conjugation in barium titanate are demonstrated, and illustrative examples of applications of phase conjugation are presented. Optical bistability is one of the most exciting phenomena occurring in the nonlinear optics of solids. In the article by B.S. Wherrett and D.C. Hutchings an authoritative treatment of optical bistability is given. In the wake of an interesting introductory section, the linear Fabry-Perot etalon and nonlinear refraction associated with electronic and thermal nonlinearities are discussed. The dynamics and the steady-state solutions of the nonlinear etalon and cavity optimisation are treated. Also switching power, power-time products, switching dynamics, and transphasor operation are described. For completeness, brief comments on a number of schemes for obtaining bistability, which are alternative to that of refractive bistability in Fabry-Perot cavities with reflective feedback, are given.

In recent years organic materials have emerged as an important class of nonlinear optical materials. As described in the article by P.N. Prasad, organic materials offer unique properties for both fundamental and applied research. The nonlinearities of the organic materials are related to the molecular structure of these systems and to their unique chemical binding. In the article by P.N. Prasad, the scope of the research on the organic materials is demonstrated by the variety of nonlinear effects one can obtain. The still very limited basic microscopic understanding of the optical nonlinearities of organic systems, and recent experimental studies are discussed. Following a section on measurements in soluble materials the dynamics of resonant third-order nonlinear processes and the role of free carriers and excitons are treated. Finally, related to organic systems, specific device possibilities, namely the optical waveguide and the nonlinear Fabry-Perot etalon are studied.

In the early days of the modern period of nonlinear optics, sum- and difference-frequency generation, harmonic generation, and parametric processes were extensively investigated. In the pedagogical paper by T. Skettrup, these effects are described in detail and it is demonstrated by results obtained from recent research related to so-called quasi phase matching, where the sign of the nonlinear susceptibility is reversed periodically with a period equal to the coherence length, that these subjects are still young and full of excitements.

[1] D. Brewster, Phil.Trans. A **105,** 60 (1815).

[2] J. Kerr, Phil.Mag. **50,** 337 (1875).

[3] W.C. Röntgen. Ann.Phys.Chem. **18,** 213 (1883).

[4] A. Kundt, Ann.Phys.Chem. **18,** 228 (1883).

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Part I

Basic Macroscopic Concepts

Second Order Nonlinear Optical Effects

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Abstract. The nonlinear optical effects of second order are discussed. The notation of nonlinear optics is reviewed. The second order susceptibility is derived from the anharmonic oscillator model. The three coupled wave equations describing three wave mixing are presented. Second harmonic generation, phase matching, parametric amplification and oscillation are finally discussed.

1. Introduction

The purpose of the present chapter is to introduce the concepts of nonlinear optics and to review different second order effects. In the first section the notation is introduced, then the second order susceptibility is derived in a purely phenomenological way using the anharmonic oscillator model. Three wave mixing is described and applied to the cases of frequency doubling and parametric amplification and oscillation.

2. Notation

When an electromagnetic field $\bar{E}(\bar{r},t)$ is applied to a material, the response of the material is a polarization $\bar{P}(\bar{r},t)$ (i.e. the induced dipole moment per unit volume) given in general by

$$\bar{P}(\bar{r},t) = \varepsilon_o \int_{-\infty}^{\infty} \int_{-\infty}^{t} \bar{\bar{\chi}}(\bar{r},\bar{r}',t-t')\, \bar{E}(\bar{r}',t')\, d^3\bar{r}'dt' \tag{1}$$

where ε_o is the vacuum permittivity and $\bar{\bar{\chi}}$ is the susceptibility tensor of the material. For systems exhibiting spatial invariance the dependence on $(\bar{r},\bar{r}')$ can be replaced by $(\bar{r}-\bar{r}')$. The truncation of the time integration is necessary since the response $\bar{P}(\bar{r},t)$ is only a function of the behaviour of the electric field in the past. This causality requirement leads to the well-known Kramers-Kronig relations /1/ between the real and imaginary parts of χ in the frequency domain. Assuming spatial invariance the Fourier transform of (1) with respect to time and space yields

$$\bar{P}(\bar{k},\omega) = \varepsilon_o\, \bar{\bar{\chi}}(\bar{k},\omega)\, \bar{E}(\bar{k},\omega)\,. \tag{2}$$

Often the local response approximation is applied (i.e. the response $\bar{P}(\bar{r},t)$ is considered to depend only on the field $\bar{E}(\bar{r},t)$ at the space coordinate $\bar{r}$). In this case (2) can be written

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$$\bar{P}(\omega) = \varepsilon_o \bar{\bar{\chi}}(\omega)\ \bar{E}(\omega) . \qquad (3)$$

If the material is nonlinear the susceptibility $\bar{\bar{\chi}}(\omega)$ can be expanded in powers of the electric field and (3) can be written

$$\bar{P}(\omega) = \varepsilon_o(\bar{\bar{\chi}}^{(1)}(\omega) \cdot \bar{E}(\omega) + \bar{\bar{\chi}}^{(2)}(\omega) : \bar{E}(\omega)\bar{E}(\omega) + ..) . \qquad (4)$$

In the literature several notations are used. Shen /2/ uses (4) directly with $\bar{E}$ and $\bar{P}$ expressed as complex quantities. This notation is simple and appealing. Yariv and Yeh /3/ use another notation with $\bar{E}$ and $\bar{P}$ expressed as real quantities. This notation is widely used and will be applied in the following. In this notation one must be careful with factors of 2 which appear, in particular, when considering harmonic generation. Following Yariv and Yeh /3/ (4) is often written in the following form:

$$P_i^r = \varepsilon_o(\chi_{ij}E_j^r + 2d_{ijk}E_j^rE_k^r + 4\chi_{ijkl}E_j^rE_k^rE_l^r + \ldots) \qquad (5)$$

where P_i^r and E_i^r are the i'th components of the field, and where summation over repeated indices is assumed. The superscript r indicates that these fields are real fields that can be expressed in terms of their complex amplitudes as follows:

$$P_i^r = \tfrac{1}{2}(P_i^{\omega} e^{i(\bar{k}\cdot\bar{r}-\omega t)} + c.c.) \quad (i = x,y,z) \qquad (6)$$

where c.c. means complex conjugate.

In the following we shall only consider second order nonlinearities, i.e. the term

$$P_i^r = 2\varepsilon_o\ d_{ijk}\ E_j^r\ E_k^r\ . \qquad (7)$$

Consider two fields at frequencies ω_1 and ω_2

$$E_j^r = \tfrac{1}{2}(E_j^{\omega_1}\ e^{i(\bar{k}_1\cdot\bar{r}-\omega_1 t)} + c.c.) \quad (j = x,y,z) \qquad (8)$$

and

$$E_k^r = \tfrac{1}{2}(E_k^{\omega_2}\ e^{i(\bar{k}_2\cdot\bar{r}-\omega_2 t)} + c.c.) \quad (k = x,y,z) . \qquad (9)$$

Using these expressions (7) becomes

$$P_i^r = 2\varepsilon_o d_{ijk}\ (\tfrac{1}{2}E_j^{\omega_1}\ e^{i(\bar{k}_1\cdot\omega_1 t)} + \tfrac{1}{2}E_j^{\omega_2}\ e^{i(\bar{k}_2\cdot\bar{r}-\omega_2 t)} + c.c.)$$

$$\times\ (\tfrac{1}{2}E_k^{\omega_1}\ e^{i(\bar{k}_1\cdot r-\omega_1 t)} + \tfrac{1}{2}E_k^{\omega_2}\ e^{i(\bar{k}_2\cdot\bar{r}-\omega_2 t)} + c.c.) . \qquad (10)$$

Due to the product in (10) terms oscillating with both $(\omega_1+\omega_2)$ and $(\omega_1-\omega_2)$ appear. Considering only the sum-frequency terms and using (6) equation (7) reduces to

$$P_i^{\omega_3=\omega_1+\omega_2} = \varepsilon_o d_{ijk}\ E_j^{\omega_1}\ E_k^{\omega_2} + \varepsilon_o d_{ijk}\ E_k^{\omega_2}\ E_j^{\omega_1} \qquad (11)$$