PART 2

MODERN NONLINEAR OPTICS

Edited by

Myron Evans
Stanisław Kielich

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Ilya Prigogine and Stuart A. Rice, Series Editors

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Part 2

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Series Editors

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INTRODUCTION

Few of us can any longer keep up with the flood of scientific literature, even in specialized subfields. Any attempt to do more and be broadly educated with respect to a large domain of science has the appearance of tilting at windmills. Yet the synthesis of ideas drawn from different subjects into new, powerful, general concepts is as valuable as ever, and the desire to remain educated persists in all scientists. This series, *Advances in Chemical Physics*, is devoted to helping the reader obtain general information about a wide variety of topics in chemical physics, a field that we interpret very broadly. Our intent is to have experts present comprehensive analyses of subjects of interest and to encourage the expression of individual points of view. We hope that this approach to the presentation of an overview of a subject will both stimulate new research and serve as a personalized learning text for beginners in a field.

ILYA PRIGOGINE
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PREFACE

Statistical molecular theories of electric, magnetic, and optical saturation phenomena developed by S. Kielich and Piekara in several papers in the late 1950s and 1960s clearly foreshadowed the developments of the next thirty years. In these volumes, we as guest editors have been honored by a positive response to our invitations from many of the most eminent contemporaries in the field of nonlinear optics. We have tried to give a comprehensive cross section of the state of the art of this subject. Volume 85 (Part 1) contains fourteen review articles by the Poznań School and associated laboratories, and volume 85 (Part 2 and Part 3) contain a selection of reviews contributed from many of the leading laboratories around the world. We thank the editors, Ilya Prigogine and Stuart A. Rice, for the opportunity to produce this topical issue.

The frequency with which the work of the Poznań School has been cited in these volumes is significant, especially considering the overwhelming societal difficulties that have faced Prof. Dr. Kielich and his School over the last forty years. Their work is notable for its unfailing rigor and accuracy of development and presentation, its accessibility to experimental testing, the systemic thoroughness of the subject matter, and the fact that it never seems to lag behind developments in the field. This achievement is all the more remarkable in the face of journal shortages and the lack of facilities that would be taken for granted in more fortunate centers of learning.

We hope that readers will agree that the contributors to these volumes have responded with readable and useful review material with which the state of nonlinear optics can be measured in the early 1990s. We believe that many of these articles have been prepared to an excellent standard. Nonlinear optics today is unrecognizably different from the same subject in the 1950s, when lasers were unheard of and linear physics ruled. In these two volumes we have been able to cover only a fraction of the enormous contemporary output in this field, and many of the best laboratories are not represented.

We hope that this topical issue will be seen as a sign of the ability of scientists all over the world to work together, despite the frailties of human society as a whole. In this respect special mention is due to Professor Mansel Davies of Criccieth in Wales, who was among the first in the West to recognize the significance of the output of the Poznań School.

MYRON W. EVANS

Charlotte, North Carolina
October 1993

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HOLOGRAPHY AND DOUBLE PHASE CONJUGATION

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CONTENTS

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I. INTRODUCTION

Two- and four-wave mixing processes in nonlinear optics have proved to be extremely useful. Several applications in optical image processing, optical communications, real-time holography, and opto-electronic neural networks have been developed in recent years. Both processes are accurately and appropriately described in terms of ordinary holography occurring in real time. The spatial or temporal interference between coherent beams spatially modulates the characteristics of the recording medium, storing information. This information can be retrieved simultaneously or later with another coherent beam. Wave mixing between mutually incoherent beams is a new phenomenon which occurs in the same materials. Double phase conjugation is one such mutually incoherent beam-coupling process, which also produces accurate three-dimensional images. Though there are some similarities between real-time holography and double phase conjugation, we will show that there are distinct differences between the two processes, requiring a new holographic interpretation of double phase conjugation. The advantage of this interpretation is that double phase conjugation can be viewed as a more general form of holography—holography with mutually incoherent sources.

This chapter is devoted to double phase-conjugate mirror (DPCM). Real-time holography and all multiwave mixing processes including mutually incoherent beam coupling (MIBC) are dynamic self-diffraction processes. We present a comprehensive review of holography (Section II), volume holography and dynamic self-diffraction (Section III), two- and four-wave mixing (Sections IV–VI), and mutually incoherent beam coupling (Section VII) to establish a link between holography and double phase conjugation. Emphasis is placed mostly on the studies with photorefractive materials, because they are the materials of choice for MIBC/DPCM and real-time holography. The double phase-conjugate mirror is reviewed in Section VIII. The holographic interpretation of the two- and four-wave mixing processes and the physical mechanism responsible for MIBC lead us to believe that DPCM is a new type of holography in which the primary holograms interact to create another hologram. This "second-order" holography is explored in Section IX. Finally, the relaxed stability and coherence requirements of the incident beams lead to new applications in optical imaging and optical communications, some of which are presented in Section X. Due to the limited scope of this chapter, the interested reader is referred to other chapters in the book and to the following for additional information: photorefractive materials and their applications,[1-3,7] volume holography,[4-6,23] two-wave mixing,[17] degenerate four-wave mixing and phase conjugation,[3,53] and photorefractive oscillators.[17,40,68]

II. HOLOGRAPHY

Holography is an optical process by which storage and retrieval of optical wavefronts is possible. Spatial information about the object is contained in the time evolution of the two-dimensional wavefronts emanating from the object. Complete information about the phase and amplitude of the wavefronts is needed to reconstruct the original object. Since recording materials do not respond to the phase directly, interferometric techniques are used to record the phase information. This places severe restrictions on the illuminating beams. For good contrast fringes and hence good efficiency, the illuminating source must have good coherence properties. High mechanical stability of the source and the object is required because of the time requirements of recording. The availability of monochromatic coherent laser sources has made possible the three-dimensional imaging of objects and has led to a large number of industrial applications. Holographic processing involves two steps: (1) recording and (2) reconstructing of the object wavefront. We review each briefly.

A. Holographic Recording

In the recording process, mutually coherent object and reference wavefronts are brought together to interfere (Fig. 1). A recording medium, such as a photographic plate, placed in the overlap region of the two wavefronts records the interference pattern.

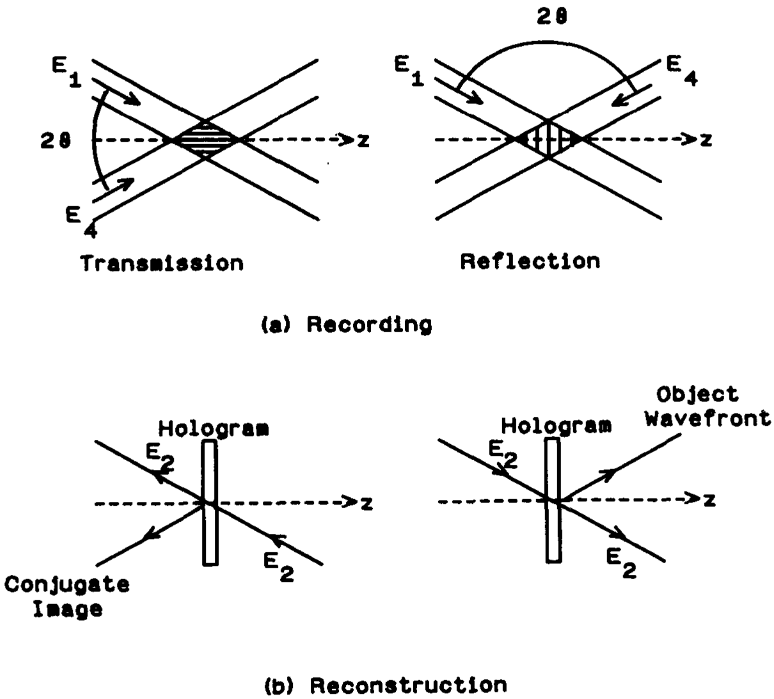


Figure 1. Holographic recording and wavefront reconstruction. E_1 and E_4 are the object and reference beams, respectively; E_2 is the reconstructing beam.

Assuming that the two beams are plane waves with parallel polarizations, the electric field of the object wavefront can be written as

$$E_1(\mathbf{r}) = A_1 \exp[-i(\omega t - \mathbf{k}_1 \cdot \mathbf{r})] \tag{1}$$

and that of the reference wavefront as

$$E_4(\mathbf{r}) = A_4 \exp[-i(\omega t - \mathbf{k}_4 \cdot \mathbf{r})] \tag{2}$$

The intensity I_T of the light in the overlapping region is given by

$$I_T = |E_1(\mathbf{r}) + E_4(\mathbf{r})|^2 \tag{3}$$

For a recording medium in the xy plane, if the direction of propagation is along the z axis, and 2ϑ is the angle between the wave vectors $\mathbf{k}_1$ and $\mathbf{k}_4$,

$$I_T = A_0^2\left[1 + m\cos\left(\frac{2\pi y}{d}\right)\right] \tag{4}$$

where m is the modulation depth given by

$$m = \frac{2A_0A_R}{A_0^2 + A_R^2} \tag{5}$$

and $d = \lambda/2\sin\vartheta$ is the spacing between the inteference maxima.

The spatial variance in the light intensity corresponding to the total field is recorded in the medium in the form of either an optical density pattern leading to an absorption grating or a phase shift pattern leading to a phase hologram. The transmission $T(y)$ of the electric vector through the absorption grating generated by I_T can be expressed as[5]

$$T(y) = T_0 + \beta E_0 m\cos Ky \tag{6}$$

where $K = 2\pi/d$ is the grating vector, T_0 is constant field transmission, E_0 is the exposure, and β is the transfer characteristic of the recording material.

In a phase grating the field transmission is given by[5]

$$T(y) \propto \left\{J_0(\phi_1) + 2\sum\left[J_n(\phi_1)\frac{\cos(nKy)}{i^n}\right]\right\} \tag{7}$$

where $J_n(\phi_1)$ are the Bessel functions of the phase modulation ϕ_1. $J_0(\phi_1)$ contributes to the constant background, while $J_1(\phi_1)$ contributes to the first-order diffracted intensity in reconstruction.

B. Wavefront Reconstruction

To reconstruct the object wavefront, we illuminate the holographic recording or hologram by a reconstruction beam E_2 (Fig. 1b). The transmitted wave-front field for the case of absorption grating is given by

$$T(y) = E_2 e^{-i\omega t}\left[T_0 e^{iky} + \tfrac{1}{2}\beta E_0 m\left[e^{-i(K-k)y} + e^{i(K+k)y}\right]\right] \tag{8}$$

The first term is a plane wave propagating in the direction of the reconstruction beam. The second term corresponds to the primary image beam. The third term corresponds to a beam traveling in the opposite direction to the object beam, and forms the conjugate image. In the case of phase gratings, reconstruction by the illuminating beam leads to the undeviated, object, and conjugate beams.

The diffraction efficiency of the hologram is defined as the ratio of the intensity of the reconstructed wavefront to the intensity of the reconstruction beam. The maximum efficiency of transmission hologram with a single-layer absorption grating is 0.0625, and that of a phase grating is 0.339. A single-layer reflective hologram is a reflection grating that can be blazed to obtain high diffraction efficiency. Reflection holograms with reconstruction efficiencies of 0.85 have been reported.

III. VOLUME HOLOGRAPHY

The efficiency of a hologram can be improved by increasing the thickness of the recording medium. This allows for more diffraction gratings to be written in the recording medium. The hologram with multiple layers of recording is called a "thick" hologram. If d is the thickness and Λ is the grating spacing, then for a thick hologram $d > n\Lambda^2/2\pi\lambda$. For sufficiently thick recording materials, the total beam overlap volume can be used to write a volume hologram. Because of the multiple planes at which light diffraction occurs in a thick hologram during reconstruction process, diffraction at or near the Bragg angle leads to efficient wave-front reconstruction. The higher diffraction orders are quenched by interference. This is true for both transmission and reflection holograms.

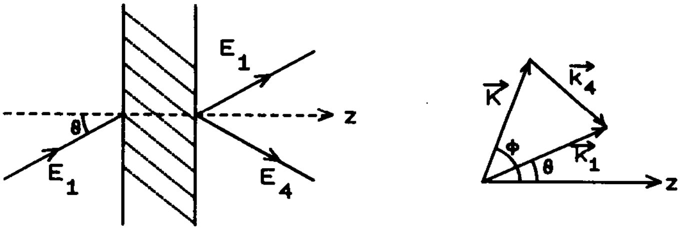
A. Thick Gratings and Beam Coupling

In thick gratings, the illuminating beam is strongly depleted as it propagates through the material due to large diffraction efficiency. Within the hologram, two mutually coherent beams are traveling: the incoming reference beam E_1 and the outgoing signal beam E_4 (Fig. 2). As the beams propagate, each beam is diffracted in the direction of the other beam at the grating surfaces. Energy exchange takes place between the beams and the beams are coupled. Kogelnik[6] analyzed these spatially dynamic interaction processes in terms of the coupled wave theory and obtained expressions for diffraction efficiencies for reflection and transmission holograms with absorption and phase gratings.

Wave propagation in the holographic grating is described by the scalar wave equation

$$\nabla^2 E + k^2 E = 0 \tag{9}$$

For small amplitudes of spatial modulations in the refractive index n_1 and



(a) (b)

Figure 2. Beam coupling in a volume hologram: (a) geometry of the interacting beams; (b) wave-vector diagram. **K** is the grating vector.

the absorption coefficient α_1, the propagation constant k in the material can be expressed in terms of the dielectric constant ε_0, the average conductivity σ_0, the grating vector **K**, and, the coupling constant κ as

$$k^2 = \beta^2 - 2\mathrm{i}\alpha\beta + 2\kappa\beta(\mathrm{e}^{\mathrm{i}\mathbf{K}\cdot\mathbf{r}} + \mathrm{e}^{-\mathrm{i}\mathbf{K}\cdot\mathbf{r}}) \tag{10}$$

where

$$\beta = \frac{2\pi\varepsilon_0^{1/2}}{\lambda} \qquad \alpha = \frac{\mu c \sigma_0}{2\varepsilon_0^{1/2}} \qquad \kappa = \frac{\pi n_1}{\lambda} - \frac{\mathrm{i}\alpha_1}{2} \tag{11}$$

κ describes the coupling between the reference beam E_1 and signal beam E_4. Amplitudes $A_1(z)$ and $A_4(z)$ vary along the propagation direction as a result of the coupling. The wave vector $\mathbf{k}_4$ is forced by the grating to satisfy the relation $\mathbf{k}_4 = \mathbf{k}_1 - \mathbf{K}$.

If the reference beam is incident at an angle ϑ with respect to the direction of propagation z, and the grating wave vector **K** is slanted at an angle ϕ, the Bragg condition is given by

$$\cos(\phi - \vartheta) = \frac{K}{2\beta} \tag{12}$$

From the scalar wave equation (9), assuming that the $A_1(z)$ and $A_4(z)$ vary slowly, the following set of coupled equations are obtained:

$$c_\mathrm{R}\frac{\mathrm{d}A_1}{\mathrm{d}z} + \alpha A_1 = -\mathrm{i}\kappa A_4 \tag{13}$$

$$c_\mathrm{S}\frac{\mathrm{d}A_4}{\mathrm{d}z} + (\alpha + \mathrm{i}\theta)A_4 = -\mathrm{i}\kappa A_1 \tag{14}$$

where $c_R = \cos\vartheta$, $c_S = \cos\vartheta - K/2\beta$, and θ is the dephasing measure given by

$$\theta = K\cos(\phi - \vartheta) - \frac{K^2\lambda}{4\pi n} \tag{15}$$

The physical picture of the diffraction process is reflected in the coupled equations (13)–(15). As the waves propagate in the material, the amplitudes of the waves change due to coupling to the other waves ($\kappa A_1, \kappa A_4$), to absorption ($\alpha A_1, \alpha A_4$), or to both. For deviation from the Bragg condition, the two beams are forced out of synchronization and the interaction decreases.

A complete analysis of the wave propagation in lossless, lossy, and slanted, transmission, and reflection holograms was given by Kogelnik.[6] The results show that the diffraction efficiencies are less for absorption gratings than for phase gratings. The maximum diffraction efficiencies are 0.037 and 0.072 for lossless, unslanted absorption gratings in transmission and reflection holograms, respectively. The grating slant improves the efficiencies of the lossy absorption gratings, though the maximum efficiencies are still below those of the lossless gratings. The maximum diffraction efficiency for lossless, unslanted phase grating is ~ 1.00 both for transmission and reflection holograms. As in the case of absorption gratings, the efficiency of the lossy phase gratings can be improved by grating slant. All these results have been verified experimentally.

One of the important applications of volume holography is in information storage in holographic memory systems. The diffraction limited storage densities attainable in volume holograms are very high. Several materials, such as photographic emulsions, photochromic glasses, dichromate sensitized gelatin, alkali halide, and electro-optical crystals, have been used in these applications. The condition for the thick holograms, $Q = 2\pi\lambda d/n\Lambda^2 \gg 1$, can be easily met for thickness d of a few micrometers. Of all these recording materials, the electro-optic crystals are of particular interest since the phase grating in these materials is phase shifted from the sinusoidal light interference pattern. The result is a strong coupling of the writing beams during the recording process. This leads to nonreciprocal energy exchange and self-diffraction of the writing beams.

B. Photorefractive Materials

The electro-optic materials in which the refractive indices are changed by light-induced electric fields are called photorefractive materials. A large number of materials have been observed to show photorefraction at moderately low illuminations; $BaTiO_3$, $Bi_{12}SiO_{20}$(BSO), $Bi_{12}GeO_{20}$(BGO), $Bi_{12}TiO_{20}$(BTO), $KNbO_3$, $LiNbO_3$, and $Sr_xBa_{1-x}Nb_2O_6$(SBN). The materials are intrinsically transparent in the visible region. But the presence of impurities permits the generation of photoexcited charges which lead to light-induced fields under nonuniform illumination. The materials are erasable and can be used for read–write applications.

The generation and transportation of charge carriers in electro-optic materials have been investigated by several workers.[7] Two charge transportation mechanisms have been proposed. In the charge hopping model,[8] the charge carriers that are excited from the donor sites in the presence of light, hop to adjacent sites with a probability proportional to the intensity of the light. The drift of the carriers by hopping continues in all directions until the carriers are out of the illuminated areas, resulting in a net electric field. In the band-conduction model[9] (Fig. 3a), the charge carriers are excited from the donor levels to the conduction band. The charges

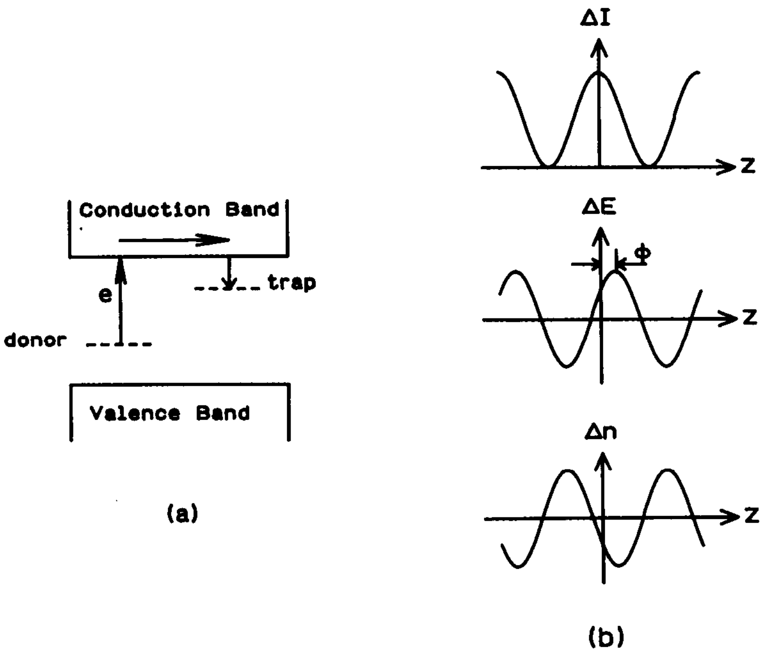


Figure 3. (a) Energy-level diagram of a photorefractive material in the band conduction model; (b) the spatial distribution of the intensity I, the space-charge field E, and the refractive index n under sinusoidal illumination. Φ is the phase shift between the light-intensity pattern and the space-charge field.