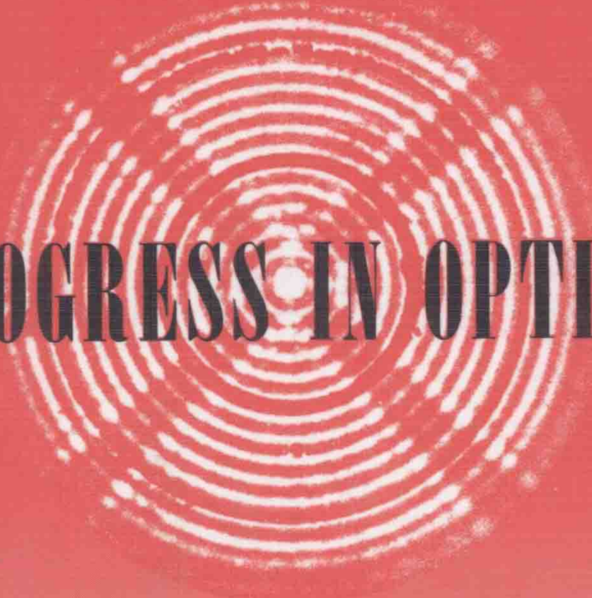
EMIL WOLF

EDITOR



PROGRESS IN OPTICS

VOLUME XXXVI

CONTRIBUTORS

L.A. APRESYAN
M. BERTERO
M. BERTOLOTTI
I. BIALYNICKI-BIRULA
V. CHUMASH
I. COJOCARU
C. DE MOL
E. FAZIO
P. HARIHARAN
YU.A. KRAVTSOV
F. MICHELOTTI
B.C. SANDERS

NORTH-HOLLAND

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EDITED BY

E. WOLF

University of Rochester, N.Y., U.S.A.

Contributors

L.A. APRESYAN, M. BERTERO, M. BERTOLOTTI, I. BIALYNICKI-BIRULA,
V. CHUMASH, I. COJOCARU, C. DE MOL, E. FAZIO,
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PREFACE

This volume presents five review articles that cover a broad range of topics which are likely to be of interest to many scientists concerned with optics and related subjects.

The first article, by V. Chumash, I. Cojocaru, F. Fazio, F. Michelotti and M. Bertolotti, deals with nonlinear optical properties of chalcogenide glasses. These materials have many interesting structural properties some of which are useful for applications to integrated active optical devices. This article presents a review of experimental measurements of nonlinear absorption coefficients and nonlinear refractive indices of such materials. A review of various models formulated to explain their properties is also included.

The second article, by P. Hariharan and B.C. Sanders, presents a review of quantum effects in optical interferometry. After a brief introduction concerning sources of nonclassical light, second- and fourth-order interference, the geometric phase, two-photon interferometry, complementarity and quantum limits are discussed. Experiments are also reviewed involving the generation of pairs of photons in entangled states, which are used to investigate some puzzling features of quantum mechanics, including tests of Bell's inequalities, quantum erasers and single-photon tunneling.

The article which follows, by M. Bertero and C. De Mol, reviews researches on super-resolution, i.e. the possibility of overcoming the classical diffraction limit of about half a wavelength. The problem is shown to be essentially equivalent to extrapolating the spatial frequency spectrum of the object beyond the spectral band of the optical system. It is demonstrated that in the presence of noise significant super-resolution can be achieved when the linear dimensions of the object are comparable with the resolution limit of the system. Some practical applications are also considered, particularly in the field of confocal scanning microscopy and in connection with inverse diffraction from far-field and near-field data.

The next article, by Yu.A. Kravtsov and L.A. Apresyan, is concerned with the theory of radiative energy transfer. The traditional theory is phenomenological, based largely on the intuitive concept of geometrical rays. More recently many attempts have been made to provide the theory with a sounder foundation. This article reviews such researches, which use the more modern techniques of wave

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The concluding article by I. Bialynicki-Birula deals with the somewhat elusive but potentially useful concept of the photon wave function. A review is presented of the century-old history of this subject. It is shown that the photon wave function bridges the gap between classical electromagnetic theory and quantum electrodynamics and it has a number of uses.

It is a pleasure to note that all the articles in this volume have been contributed by leading experts in the various fields.

Emil Wolf

Department of Physics and Astronomy
University of Rochester
Rochester, New York 14627, USA

October 1996

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I

NONLINEAR PROPAGATION OF STRONG LASER PULSES IN CHALCOGENIDE GLASS FILMS

BY

V. CHUMASH AND I. COJOCARU

Center of Optoelectronics, I.A.P., Academy of Sciences of Moldova, Academiei str. 1, Chisinau, 277028, Moldova

AND

E. FAZIO, F. MICHELOTTI AND M. BERTOLOTTI

Università degli Studi di Roma "La Sapienza", Dipartimento di Energetica, GNEQP of CNR and INFM, Via A. Scarpa 16, 00161 Roma, Italy

CONTENTS

§ 1. Introduction

Nonlinear optical effects in semiconductors are studied less frequently in the amorphous than in the crystalline state. The chalcogenide glass semiconductor (ChG) is a material that is highly resistive to crystallization. Irradiation of light could cause a change in its optical properties that is not related to the crystalline–amorphous type of transformation.

It is well known that the optical properties of ChG near the equilibrium state are primarily determined by the spectrum of the localized states in the band gap, and by their carrier concentration. Most investigations do not address the question as to how the process of carrier excitation by light takes place, however, and how these carriers relax later, especially from extended into localized states. An understanding of the relaxation processes is a basic problem in amorphous-state physics, since they represent the first step for the localization process, the basic property of ChG. Furthermore, the knowledge of the kinetics peculiarities of the electron–hole pair relaxation and localization permits the testing of different theoretical models and hypotheses (e.g., multiple carrier trapping or the assumption of high carrier mobility in extended states). Progress in clarifying these processes greatly depends on the possibility of a quantitative investigation of the spectrum of the elementary excitations in a wide energy region, with time resolution of the order of the characteristic time of the investigated elementary processes. These requests widen when the elementary excitations in a light field (external driving force) are investigated, including the nonlinear and nonstationary medium behavior in states far from the thermodynamic equilibrium.

Amorphous semiconductors, including ChG, are attractive candidates for the fabrication of all-optical passive and active devices. In recent years a variety of both passive (fibers, planar waveguides, lenses, gratings) and active (nonlinear devices mainly based on Fabry–Perot interference, optical bistability and optical hysteresis) elements have been demonstrated (Andriesh, Bykovskii, Kolomeiko, Makovkin, Smirnov and Shmal'ko [1977], Andriesh, Bykovskii, Smirnov, Cernii and Shmal'ko [1978], Suhara, Handa, Nishihara and Koyama [1982], Hajto and Janossy [1983], Andriesh, Enaki, Cojocaru, Ostafeichuk, Cerbari and Chumash [1988], Haro-Poniatowski, Fernandez Guasti, Mendez and Balkanski [1989]), Nasu, Kubodera, Kobayashi, Nakamura and Kamiya [1990], Heo, Sanghera and Mackenzie [1991], Bertolotti, Chumash, Fazio, Ferrari and Sibilia [1991], Andriesh, Chumash, Cojocaru and Enaki [1991], Asobe, Suzuki, Kanamori and Kubodera [1992], Bertolotti, Chumash, Fazio, Ferrari and Sibilia [1993], Chumash, Cojocaru, Bostan, Cerbari and Andriesh [1994].

The aim of this work is to illustrate the present state of knowledge and some unresolved problems of the nonlinear interaction of a strong laser radiation with ChG. The primary focus is nonlinear phenomena that are characteristic of the amorphous semiconductors and that, as a rule, have no analog in crystalline phase. Permanent photoinduced effects that result from laser excitation are not considered here (i.e., effects that remain in ChG after irradiation) because of previous work (see, e.g., Kastner [1985], Elliot [1986], Tanaka [1990] and the references therein).

§ 2 examines the peculiarities of the nonlinear transmission of CW laser radiation through thin ChG films. § 3 addresses nonlinear absorption of laser pulses into ChG films, and § 4 discusses optical hysteresis and nonlinear interaction of short laser pulses with the ChG. A physical model, taking into account the light interaction with nonequilibrium phonons, is considered in § 5 in order to explain the experimental results. The results of the numerical calculations of the phenomenological equations are compared with the experimental data. § 6 examines results of the Z-Scan spectroscopy investigation of ChG thin films under interband and intraband CW and picosecond irradiation. Nonlinear refraction, nonlinear absorption, and permanent photostructural changes on ChG thin films, suitable for planar waveguiding structures, are reviewed and possible applications of ChG refractive index changes are discussed.

§ 2. Nonlinear Transmission of CW Laser Radiation through Thin ChG Films

Several researchers studied some of the light-induced reversible changes of the ChG optical constants to examine the nonlinear transmission of focused CW laser radiation through thin film samples.

Toth, Hajto and Zentai [1977] observed a nonlinear change of the light transmission, when $GeSe_2$ and AsSe films were irradiated with a focused Ar-laser beam. With the purpose of excluding the contribution of irreversible changes of the optical parameters, due to photostructural changes, the samples were "stabilized" in the laser beam in advance or were annealed. The properties of the reversible nonlinear change of the ChG sample transparency depend on the laser input intensity.

The light transmission of ChG samples decreases (laser radiation with intensity from 2.5 W/cm^2 up to 15 W/cm^2) with the irradiation time (t) according to a relation close to t^{-1}, whereas the value of the relative transmission change is almost proportional to the intensity of the exciting light. For example, when the Ar-laser exciting intensity (with wavelengths $\lambda_{1ex} = 4880$ Å and $\lambda_{2ex} = 5145$ Å) increased from 0 to 15 W/cm^2, the light transmission of an AsSe film (with $E_g = 1.86$ eV and thickness $d = 1.85$ μm) decreased 5 times, whereas the transmission of a $GeSe_2$ film ($E_g = 2.1$ eV, $d = 6.4$ μm) decreased 2.4 times. After switching off the exciting light the ChG film transmission recovers its starting value exponentially. The transmission changes of the ChG films occur with characteristic times of several seconds and no fast components are revealed (Toth, Hajto and Zentai [1977]). It is worth noting that, as a result of the ChG photostructural changes, the studied films are bleached ($GeSe_2$) or darkened (AsSe); in contrast, in an intensive CW laser radiation their transmission always decreases.

Some studies (Hajto, Zentai and Kosa Somogyi [1977], Hajto and Janossy [1983]) reported that with a CW-focused He–Ne laser ($\lambda = 632.8$ nm, with a fixed radiation intensity), the photocurrent, transmission, and reflection coefficients of $GeSe_2$ films (deposited on glass substrates or self-supported in the air) show periodic oscillations in time. The material returns to its initial transparent state if the laser is switched off. The interaction of the laser radiation with the air self-supported $GeSe_2$ thin films takes place at a considerably smaller light intensity (~40–50 W/cm^2), compared with the films on glass substrates (~2 kW/cm^2). The frequency (3–50 Hz) and amplitude of the light transmission oscillations noticeably depend on the incident radiation intensity: an increase in the laser radiation intensity is followed by an increase in the amplitude of the transmission oscillations and by a decrease of their frequency (fig. 1). It should be noted that the transmission oscillations are observed in strictly limited ranges of the laser intensities (from 1.39 kW/cm^2 up to 2.65 kW/cm^2 for the $GeSe_2$ film on the glass substrate). Near the laser threshold intensity the detected oscillations are distinguished by a high stability, and after about 10^4 cycles of oscillations no change in the ChG structure or any sign of matter transport are detected. A logarithmic time dependence of the amplitude and frequency of the oscillations was reported by Hajto, Janossy and Choi [1985]. It was not possible to find an oscillation regime of the light transmission in crystalline $GeSe_2$, indicating that the clue to understand the physical mechanism lying at its base is related to the amorphous nature of the ChG.

Another kind of nonlinear interaction of laser radiation with the ChG, which has been revealed as an optical bistable light transmission, was found for the