Photoinduced Nonlinear Optical Phenomena in Glasses and Optical Fibers

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ABSTRACT

The combination of nonlinear optics and photoinduced effects in glasses and optical fibers gives rise to interesting new optical phenomena. These phenomena, which include photoinduced refractive index gratings and second harmonic generation in doped silica fibers, are reviewed and their mechanisms, the roles of defects and dopants, and applications are discussed.

I. INTRODUCTION

Glasses are used as optical materials for a wide variety of applications ranging from lenses to optical fibers. In most of these applications the glass serves as a transmission medium with its optical properties being described by the absorption coefficient and the refractive index, which are contained in the real and imaginary part of the linear optical susceptibility. When the light intensities are relatively low, which is usually the case for irradiation with a lamp or low power cw laser, the linear susceptibility is sufficient to describe the optical properties. However, above a certain material-dependent intensity, processes that depend nonlinearly on the light intensity start to become important and nonlinear optical phenomena can be observed. Examples of such processes are second and third harmonic generation, two-photon absorption, intensity-dependent refractive index effects and stimulated Raman and Brillouin scattering [1,2].

A special class of optical processes are those in which the action of the light changes the electrical, structural or optical properties of the material permanently or at least for times much longer than those normally associated with excitation and emission processes. A well-known example of such a process is the photodarkening effect, i.e. the increase of absorption upon irradiation, which occurs, for instance, in Mn^{2+} doped glasses [3].

In the above example the photoinduced change can be achieved through a one-photon excitation process. Nonlinear optical processes can induce similar permanent changes in the optical properties of the glass and it is this combination of nonlinear optics and photoinduced changes that will be discussed in this paper. The effects that have been

observed are 1) photoinduced refractive index gratings in rare-earth doped bulk glasses 2) photoinduced refractive index gratings in optical fibers and 3) photoinduced second harmonic generation in optical fibers. Although the origin of these effects is not understood in detail the presence of defects or dopants in the glass has been shown to play an important role.

The observation of nonlinear optical effects is in general made easier with high intensities and long optical interaction lengths. These conditions are favored in optical fibers and this is the reason why most nonlinear optical effects, including the photoinduced nonlinearities, are much more easily observed in optical fibers than in bulk glasses. In this paper we will review the phenomena observed in optical fibers. An account of the refractive index gratings in rare-earth doped bulk glasses can be found in refs. 4-8.

II. NONLINEAR OPTICS

Many excellent monographs on nonlinear optics are available [e.g. 1,2], so here the relevant concepts and phenomena will be presented very briefly. When light, with an electrical field vector E, interacts with a medium a polarization, P, is induced in the medium. This polarization itself acts as a source of electromagnetic radiation and thereby modifies the optical field, resulting in an output which is different from the input. The output optical field can be found using Maxwell's equations to obtain a set of coupled wave equations, containing both E and P. The relation between E and P is in general given by:

$$P=\chi^{(1)}.E_1+\chi^{(2)}:E_1E_2+\chi^{(3)}:E_1E_2E_3+.... \quad (1)$$

with

$$P^{(1)}(k,\omega)=\chi^{(1)}(k,\omega)E(k,\omega) \quad (2)$$

$$P^{(2)}(k,\omega)=\chi^{(2)}(k=k_1+k_2,\omega=\omega_1+\omega_2):E(k_1,\omega_1)E(k_2,\omega_2) \quad (3)$$

$$P^{(3)}(k,\omega)=\chi^{(3)}(k=k_1+k_2+k_3,\omega=\omega_1+\omega_2+\omega_3):E(k_1,\omega_1)E(k_2,\omega_2)E(k_3,\omega_3) \quad (4)$$

Under low intensity conditions only the first linear term in this expression has to be taken into account. In this case the dependence of P on E is given by the linear optical susceptibility, $\chi^{(1)}$, which is a complex, frequency-dependent quantity that characterizes the optical properties of the medium. Under higher intensities the higher-order terms in eq. 1 become increasingly important. The higher order susceptibilities are again completely determined by the medium, but with a more complicated frequency dependence. As a result each of these higher-order terms can give rise to a variety of optical phenomena. The second-order term (eq.2) can lead to second harmonic generation (SHG) ($\omega_1=\omega_2,\omega=2\omega_1$), optical rectification ($\omega_1=-\omega_2,\omega=0$), electrooptic effect ($\omega_2=0,\omega=\omega_1$), and sum- and difference-frequency generation. The

third-order term is responsible for processes such as third harmonic generation ($\omega_1 = \omega_2 = \omega_3, \omega = 3\omega_1$), intensity dependent refractive index ($\omega_1 = -\omega_2 = \omega_3, \omega = \omega_1$), four-wave mixing etc.

Because of symmetry considerations $\chi^{(2)} = 0$ in materials with inversion symmetry. Although on a local scale the glass lacks inversion symmetry, on a scale corresponding to optical wavelengths a glass is an isotropic material and therefore expected to have $\chi^{(2)} = 0$, as is normally observed. Materials with non-zero $\chi^{(2)}$ are crystalline ferroelectrics, such as $BaTiO_3$, KTP, $LiNbO_3$ etc. In these crystalline materials SHG is observed.

For efficient conversion of the fundamental beam at ω to the second-harmonic at 2ω, a second condition, in addition to a non-zero $\chi^{(2)}$, has to be fulfilled, namely that of phasematching. The phasematching condition requires that the fundamental and second harmonic beam stay in phase with each other while passing through the material. Due to the dispersion in the material the two beams will in general get out of phase after a relatively short distance, the coherence length. Several methods, which will not be discussed here, can be employed to obtain phasematching over distances much longer than the normal coherence length [2].

III. PHOTOINDUCED REFRACTIVE INDEX GRATINGS IN OPTICAL FIBERS

In 1978 Hill et al. observed peculiar behavior in the transmission of a Ge-doped silica fiber while irradiating it with a narrowband cw argon laser at 488 nm [9,10]. The transmission of the fiber decreased as a function of time whereas at the same time the reflectivity of the fiber increased. After several minutes the reflectivity saturated at a value close to 100%. Various experiments [9-13] have shown that the observed increase in reflectivity is due to the formation of a permanent refractive index grating in the fiber which acts as a Bragg grating for the incident light, i.e. the grating spacing is half the wavelength of the light (Fig. 1a).

The process in the fiber can be described as follows. Initially a standing wave pattern is set up in the fiber by the transmitted beam and the counterpropagating beam which results from the Fresnel reflection at the fiber end. This intensity grating leads to a permanent photoinduced modulation of the refractive index. The induced index change is typically 10^{-5} to 10^{-4} for laser powers around 100 mW [12,14]. After its formation the grating appears to be permanent at room temperature. The efficiency of grating formation increases with increasing Ge concentration in the fiber; gratings are not observed in fibers that have a pure silica core [15]. The change in refractive index is believed to be the result of a complicated interplay between different types of Ge defects in the glass [16]: two-photon absorption of 488 nm light by so-called 'GeO' defects, which occur in oxygen-deficient germanosilicates [17-19], results in the release of electrons, which are subsequently trapped at other types of defect sites. The resulting change in relative defect concentrations is responsible for the observed change in refractive index. Although such a model qualitatively explains the photoinduced

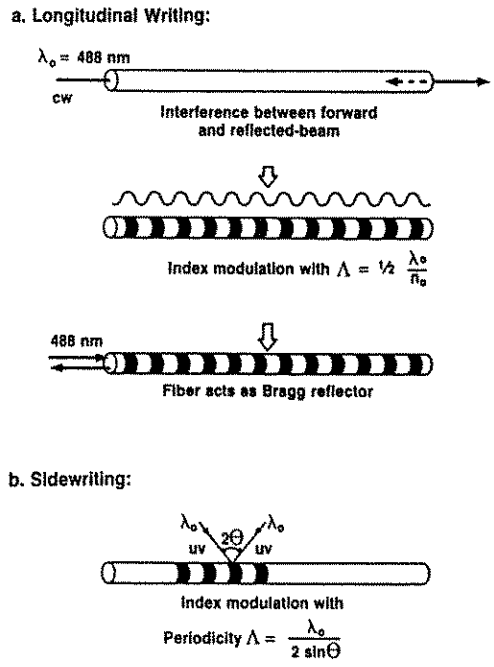

Fig. 1 Schematic diagram of the generation of a photoinduced refractive index grating in a fiber.

refractive index changes, a detailed understanding of the structure and role of the defects involved is still lacking.

Simultaneous with the discovery of the refractive index gratings, Hill et al. [9] showed that they can be used as extremely narrow bandwidth filters. One of the drawbacks of these gratings is that the wavelength that is reflected is determined by the wavelength of the laser light with which the grating is written. Since the writing wavelength in turn has to lie within the two-photon absorption band of the 'GeO' defect, the spectral range is limited. A large practical improvement in this respect was made by Meltz and coworkers [20,21] who showed that gratings could be formed by side-illumination with uv light. In this case the fiber is exposed to a two-beam interference pattern which is generated by two coherent overlapping uv laser beams. The photoinduced change now takes place via direct one-photon excitation of the 'GeO' defect and is therefore more

IV. PHOTOINDUCED SECOND HARMONIC GENERATION IN GLASS FIBERS

As discussed in section II the process of SHG is normally forbidden in a glass. In 1987, however, Margulis and Osterberg [28,29] reported that a Ge-P doped fiber that had been irradiated for several hours with a very intense laser beam at 1.06 μm (mode-locked, Q-switched, Nd-YAG laser with a peak power of 50 kW) showed a strong SH signal which grew exponentially with the time of irradiation and saturated after about 12 hours. The SH conversion efficiency was as high as 5% and the SH beam intense enough to pump a dye laser. Later Stolen and Tom [30] found that the time necessary to induce the SHG in the fiber could be substantially reduced when in addition to the very strong ir beam the fiber was simultaneously exposed to a weak green beam at 532 nm, obtained by doubling part of the ir light with a doubling crystal. This green beam is only present during the preparation process (seeding); during probing of the SHG only the ir beam is coupled into the fiber. A schematic summary of the seeded SHG process is presented in Figure 2. The initial observations of Osterberg and Margulis, referred to as self-seeding, can be considered as a special case of the seeded scenario: during irradiation with ir light alone a very small amount of green light is generated through some other, non-electric-dipole, process. This green light can then serve as a seed and together with the ir light induce SHG. In either the seeded or self-seeded case the

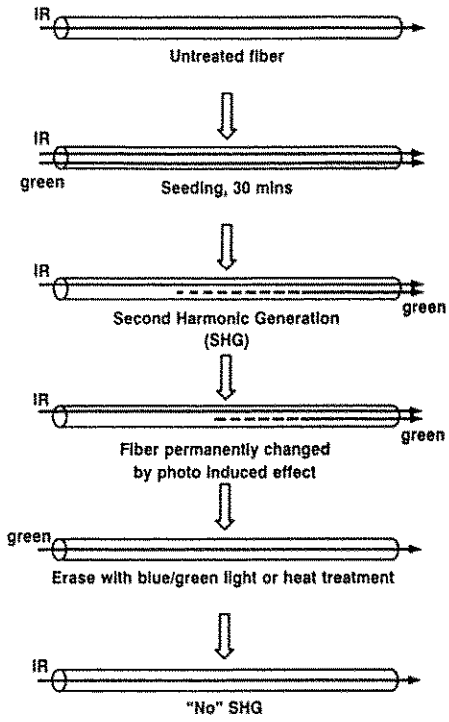

Fig. 2 Schematic diagram of the processes associated with photoinduced SHG in fibers.

optical preparation process results in a permanent change in the fiber [31], i.e. after the fiber has been stored in the dark indefinitely (weeks to months) SHG is instaneously observed when the fiber is reexposed to ir light. The induced effect can be erased by heating the fiber to $\approx$500 °C or optically by irradiating the fiber with green light alone [32].

From the above observations two conclusions can be derived: a) the optical preparation process induces a $\chi^{(2)}$ in the glass and b) because rather high conversion efficiencies for SHG can be observed in irradiated fibers, the SHG process must be phase matched, that is, the second harmonic beam must remain in phase with the input beam through a considerable length of the fiber (10-30 cm is the active length for SHG as determined by cut-back experiments [29]). Because of the difference in refractive index at 1.06 and 0.532 μm, the green beam would normally be out of phase with the ir beam after traveling through the fiber over a distance $L = (2k_\omega - k_{2\omega})^{-1}$, where k_ω and $k_{2\omega}$ are the wavevectors at ω and 2ω, respectively. L is typically 50 μm for a silica glass. However, if the $\chi^{(2)}$ in the glass is periodic with a periodicity exactly equal to L, then the green and ir beams will stay in phase. How such a periodic $\chi^{(2)}$ grating can be induced will be discussed below.

Since the discovery of SHG in fibers many theoretical models [see e.g 31, 33-37] have been suggested to explain this unexpected phenomenon. Despite the many differences between all these models, especially as far as their microscopic interpretation is concerned, the following phenomenological description is common to most of them (cf.

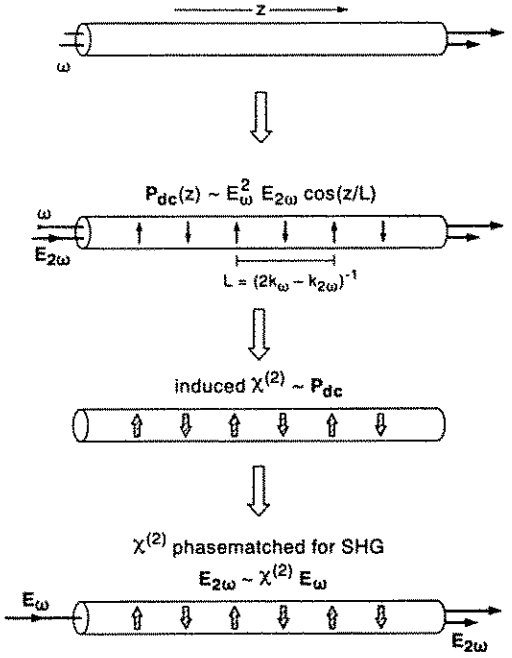

Fig. 3 Schematic diagram of the phenomenological model for generating a photoinduced SH grating in a fiber.

fig. 3). A dc polarization is induced in the fiber through a specific, model-dependent, third-order nonlinear optical process, which is allowed even in an unirradiated fiber. A permanent $\chi^{(2)}$ proportional to this dc polarization is then induced in the glass. Microscopically this $\chi^{(2)}$ can in principle be the result of alignment of dipoles in the optically induced dc field or the generation of a space-charge field. Because the periodicity of the $\chi^{(2)}$ grating, L, is determined by the nonlinear interaction between the preparation beams at ω and 2ω, subsequent probing with the fundamental beam at ω results in SHG which is automatically phasematched.

As with the photoinduced refractive index gratings, dopants or defects play a crucial role in the process of photoinduced SHG. Most of the studies have been done on Ge- or Ge/P-doped silica fibers [31], but SHG has also been reported for other systems, including semiconductor-doped glasses [38] and rare earth doped aluminosilicate glass fibers [39,40]. The effect is not observed in pure silica core fibers. Although the precise role of the defects is not understood, a number of experiments indicate the importance of photoexcitation of the defects as part of the mechanism. In one of the experiments, rather than generating the dc-field optically, it is applied externally. With fields of several thousands of Volts/cm, induced in Ge/P doped silica preforms and fibers, no permanent $\chi^{(2)}$ could be obtained [41,42]. However, when a fiber was irradiated with blue laser light during the application of the dc field a permanent $\chi^{(2)}$ was observed [43-45]. Recent work by Carvalho et al. [46], which shows an enhancement of the induced $\chi^{(2)}$ when uv light is present during seeding, also indicates the importance of metastable excited states. Other results that point to the role of Ge-defects are the observation that the photoinduced SHG increased when the fiber was exposed to hydrogen [46] and the fact that the ESR signal from induced Ge E' centers was correlated with the SHG conversion efficiency [47].

A microscopic model that can explain many of the experimental observations is based on the coherent photovoltaic effect [34,35,37]. In this model a space charge field (E_{dc}), which induces a $\chi^{(2)}$ via $\chi^{(2)} = \chi^{(3)} E_{dc}$, is generated in the glass through the asymmetric photoionization of defects. The asymmetric photoionization is the result of a quantum interference between a two-photon ionization at ω and a one-photon ionization at 2ω or higher-order interference terms. In this picture the role of the dopants is to provide defects that can be photoexcited and photoionized. Although certain features of this model have been tested experimentally [48,49], a precise understanding of the optical processes that are taking place is lacking.

As far as applications of photoinduced SHG in fibers are concerned, the conversion efficiencies are at present too low to make these fibers interesting devices. The highest conversion efficiency suggested is 13 % for 950 W infrared peak power [50], which is much lower than efficiencies obtained in single crystals (>50 %). However, a better understanding of the processes and the nature of the defects involved in the photoinduced SHG gratings could potentially lead to the design of materials with much higher efficiencies.

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和文抄録

ガラスや光ファイバー中における光誘起非線形光学効果

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ガラスは光学材料として幅広い用途に利用されているが，多くの用途は透過媒体としてであり，それらの性質は線形光学感受率の実部と虚部に含まれる，屈折率や吸収によって説明される光学的性質である．一方レーザの様な光の強度が強い場合には非線形光学現象が観測される．それらは，1)希土類ドープバルクガラスにおける光誘起屈折率グレーティング，2)光ファイバー中の光誘起屈折率グレーティング，3)光ファイバー中の光誘起第2高調波発生である．

これらの現象は，高い光強度と大きな相互作用長によって起こりやすくなるため，光ファイバー中で最もよく観測される．ガラスは局所スケールでは反転対称中心を欠いているが，光の波長に相当するスケールでは本来，等方的物質であり，従って通常観測されるように $\chi^{(2)}=0$ である．一方，基本波 ω の光を効率よく 2ω に変換するためには $\chi^{(2)}$ がゼロでないと同時に位相整合の条件が満たされねばならない．

Hill らにより，1978年に Ge-ドープシリカファイバーで光誘起屈折率グレーティング現象が発見された．即ち進行波と反射波によって定常波が生成し，これが永続的な屈折率の光誘起変調を引き起こす(図1a)．定性的なモデルとして，'GeO' 欠陥の関与説があり，この欠陥による 488 nm 光の2光子吸収により，電子が解放され，引続き他の欠陥種にトラップされるために屈折率変化が引き起こされる．この現象を利用して，Melz らは紫外光の横方向照射角によって波長可変の超狭幅フィルターができることを示した(図1b)．

SHG については1987年のファイバー中での発見以来，数多くの理論モデルが提唱されている．これまでの SHG の観測は Ge，Ge-P ドープシリカファイバー，あるいは半導体ドープガラスや希土類ドープファイバーにおけるもので，純粋シリカコアファイバー中ではこの効果は観測されていない．これらの原因は詳細は明らかにはなっていないが，欠陥あるいはドーパントの存在が重要な役割を果たしていることが示されてきている．微視的な解釈に関する限り，以下の現象論的説明が殆どのものについて共通である．；ファイバー中に直流分極が，3次の非線形光学過程を通じて誘起される．；この直流分極に比例する永続的な $\chi^{(2)}$ がガラス中に誘起される．微視的にはこの $\chi^{(2)}$ の形成は，光誘起によって生じた直流電場における双極子の整列あるいは，空間電荷場の生成の結果であろう．；$\chi^{(2)}$ グレーティングの周期 L は基本波と2倍波の間の非線形相互作用に依って決定されるので，引き続いて起こる，基本波との作用が，自動的に位相整合された SHG を生じる(図2，3)．欠陥やドーパントは，機構の一部として，光励起あるいは光イオン化される欠陥を提供する役割をしていることが，数多くの実験により示されている．が，実際に起こっている光学過程についてはまだ詳細には理解されていない．

現在のところ変換効率は実用的なデバイスになるまでには達しておらず，これまでの最高効率は 950 W のピークパワーで 13%で，これは単結晶(50%)よりはるかに低い．しかし，光誘起 SHG グレーティングにおける過程と欠陥の本質を理解することにより，より高い効率を有する材料の設計ができる可能性がある．

(京都大学総合人間学部　田部 勢津久)