# OPTICAL NONLINEARITY IN GLASSES

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# ガラスの非線形光学特性

### [概要]

近年、酸化物ガラス、ハライドガラス、カルコゲナイドガラス、半導体微結晶、有機染料、金属粒子等をドープしたガラス等の、そしてバルク材料、光ファイバー、導波路など各種形状の材料の非線形光学現象について研究が行われている。

これらガラスの組成やドーパントの選択により、その非線形光学特性を、高出力レーザーのビームブレイクアップの制御、すべてが光デバイスによるスィッチや論理回路、極短パルスの発生、超長距離のソリトン伝送など広い応用に適するように調節することができる。

ここ数年来、高い非線形係数をもったガラスを目指して競争が展開されてきた。

この報告で、最近の非線形光学部品の研究成果と実用的なシステムに応用するのに解決しなければならない厳しい制約について述べる。

Nonlinear optical phenomena have been investigated in recent years in materials ranging from oxide, halide, and chalcogenide glasses to glasses doped with semiconductor microcrystals, organic dyes, and metal particles and in forms varying from bulk materials to optical fibers and waveguides. This selection of glass compositions and dopants provides the ability to tailor the nonlinear optical properties of glass for such diverse applications as controlling beam breakup in high-power lasers, demonstration of all-optical devices for switching and logic, ultrashort-pulse generation, and ultralong-distance soliton transmission. The past few years have witnessed a race for the highest nonlinear index coefficient in glasses. In this paper we report on the recent achievements and severe constraints that practical systems placed on the nonlinear optical components.

## INTRODUCTION

Glass is an excellent linear optical material producible with high optical quality in both bulk and fiber form. In recent years, nonlinear properties of glasses have also attracted much attention, as researchers are considering various all-optical devices that require reliable and efficient nonlinear optical materials. Many of these devices are based on fiber or planar waveguide technologies, both particularly compatible with glass fabrication techniques.

In treating nonlinear optical phenomena in glasses we make a distinction between intrinsic and extrinsic effects. For intrinsic effects the structure and composition of the glass are of primary importance in controlling the nonlinearity, whereas for extrinsic dopant effects the influence of the glass matrix is secondary in nature. Another distinction that has to be considered is whether the light frequency  $\omega$  is resonant or nonresonant with an electronic transition in the material. While resonant (absorbing) nonlinearities are large, the penalties are higher power dissipation and slower response.

They are many recent extensive summaries and reviews of these multifaceted developments (1 - 3). In this review the emphasis is on the new experimental and modeling results in the homogeneous glasses where the glass itself is responsible for the nonlinearity.

#### NONLINEAR OPTICAL PHENOMENA

The interaction of an electromagnetic field with an atomic system has been reviewed extensively with emphasis on nonlinear optics in, for example, books by Yariv (4), Shen (5) and Boyd (6). The polarization P induced in a medium by external optical electric fields E of frequency  $\omega$  is expressed by the power series

$$P(\omega) = \chi^{(1)}(\omega)E(\omega) + \chi^{(2)}(\omega = \omega_1 + \omega_2)E(\omega_1)E(\omega_2) + \chi^{(3)}(\omega = \omega_1 + \omega_2 + \omega_3)E(\omega_1)E(\omega_2)E(\omega_3) + ...., \tag{1}$$

where  $\chi^{(n)}$ , the complex dielectric susceptibilities, are tensors of rank (n + 1) and are related to the microscopic (electronic and nuclear) structure of the material (5). The various processes arising from the terms in the expansion in Eq.(1) must also conserve momentum and wavevector k.

Classical or linear optics are concerned with the first-order linear term  $\chi^{(1)}$ . The real part of  $\chi^{(1)}$  gives rise to the linear refractive index  $n_o$ ; absorption is proportional to the imaginary part of  $\chi^{(1)}$ . The higher-order terms in Eq. (1) describe nonlinear interactions. These are usually classified according to the order of the term in the series, i.e. second-order effects ( $\chi^{(2)}$ ), third order-effects ( $\chi^{(3)}$ ), etc. Within the same order, different nonlinear effects can be classified according to the number of externally applied fields and their frequencies.

# Second-Order Optical Nonlinearities - $\chi^{(2)}$

The second-order nonlinear susceptibility  $\chi^{(2)}$  is the origin of second-harmonic generation [described by  $\chi^{(2)}(-2\omega,\omega,\omega)$ ], parametric mixing  $[\chi^{(2)}(-\omega_1\pm\omega_2,\omega_1,\omega_2)]$ , and the Pockels, or electrooptic effect  $[\chi^{(2)}(-\omega,\omega,o)]$ .

The second-order nonlinearity  $\chi^{(2)}$  is important in materials having no center of inversion which is typical for some crystals, for example, LiNbO<sub>3</sub> (7) or KTP (8). In isotropic materials that possess inversion symmetry such as glasses, the second-order contributions vanish. Thus the discovery of efficient second harmonic generation in optical fibers was surprising. The mechanism responsible for this unexpected nonlinear effect in a glass has been reviewed by Krol (9). Except for this anomaly, the most important nonlinear effects in glasses are of the third order.

# Third-Order Optical Nonlinearities - $\chi^{(3)}$

The third-order contributions to the total polarization are given by

$$\chi_{i}^{(3)}(\omega_{4}) = \sum_{ikl} \chi_{jkl}^{(3)}(-\omega_{4,}\omega_{1},\omega_{2},\omega_{3}) E_{j}(\omega_{1)} E_{k}(\omega_{2}) E_{l}(\omega_{3}), \tag{2}$$

where  $\omega_4 = \omega_1 + \omega_2 + \omega_3$  and  $E_j E_k E_l$  are three separately applied electric fields each having its own frequency  $(\omega_1, \omega_2, \omega_3)$  and polarization direction (5). However, some or all of these fields may be identical. In the case of third-order nonlinearities, many different processes are possible depending on the number of applied fields and their frequencies. Processes involving  $\chi^{(3)}$  are third-harmonic generation [  $\chi^{(3)}(-3\omega,\omega,\omega,\omega)$  ], Raman and Brillouin scattering [  $\chi^{(3)}(-\omega_1,\omega_2,-\omega_2,\omega_1)$ ], and a large number of three- and four-wave mixing processes, the most common of which is degenerate four-wave mixing [ $\chi^{(3)}(-\omega,\omega,\omega,-\omega)$ ].

Perhaps the most important third-order effect from an application point of view is the optical Kerr effect, which is a four-wave mixing process and results in an intensity-dependent contribution to the refractive index. In the simplest case, a single field is applied and it changes the index of refraction of the medium according to its intensity. The total index of refraction is given by

$$n = n_0 + n_2 < E^2 >$$
, (3)

where  $n_0$  is the linear index of refraction,  $\langle E^2 \rangle$  is the time-averaged square of the electric field of the incident optical beam in esu, and  $n_2$  is the nonlinear refractive index. In SI units

$$n = n_0 + n_2 I, \tag{4}$$

where I is the intensity  $(m^2/W)$  of the incident beam. The relationship between the nonlinear index in SI and esu units is described in details elsewhere (6).

Another third-order effect is two-photon absorption. As in linear optics, it is the imaginary part of  $\chi^{(3)}$  that contributes to (nonlinear two-photon) absorption. In this case, a beam of intensity I propagating in the z-direction is attenuated as

$$\frac{\mathrm{d}\mathbf{I}}{\mathrm{d}z} = \alpha \mathbf{I} - \beta \mathbf{I}^2,\tag{5}$$

where  $\alpha$  is the linear absorption coefficient and  $\beta$  is the two-photon absorption coefficient. The specific linear combinations of  $\chi^{(3)}$  components that define  $n_2$  and  $\beta$  are dependent on the incident beam polarization and the material anisotropy and are given in the literature, for example in Ref. 10.

#### Oxide Glasses

The approaches to calculating the nonlinear optical properties of solids vary in their degree of sophistication from the rigorous to the empirical. First principles calculations of the optical properties of complex, multicomponent glasses are hampered by our lack of knowledge of the detailed local structure and resultant fields, and their variation from site to site in a glass (11). Nevertheless, recent overlapping bond-orbital calculations of the nonlinear dielectric response of crystals have increased our understanding of the relationship between structure and nonlinear behaviour (12). Rather than separating the dielectric response into additive ionic constituents, this approach emphasizes the bond nature of the phenomena. In these calculations the influence of cationic d-orbitals on electronic polarizability is explicitly recognized. Calculations now show that the largest nonlinear responses are generated by highest valence metal cations or by maximum p-d

orbital overlap (13). The d-orbital contributions to the linear and nonlinear response become dominant when bond lengths d is less than 2.0 A. From these encouraging results it is anticipated that a bond orbital theory will eventually lead to a global semiquantitative representation of the optical properties as functions of formal valency, bond length and ionic radii of the glass constituents.

The validity of the bond-orbital theory extended to glasses containing transition metal oxides was demonstrated for the first time (14). The nonlinear indices calculated by taking into account long and short Ti-O distances that are present in tetrahedral or octahedral coordination of Ti showed very good agreement with the experimentally measured nonlinear refractive indices. The calculated data were compared to measurements in  $Na_2O-TiO_2-P_2O_5$  and  $Na_2O-TiO_2-SiO_2$  glass systems.

To date, however, most progress in the prediction of third-order nonlinearities in glasses has been achieved by semi-empirical approaches. These methods, based on various assumptions and observed trends, use the analysis of the experimental data to extrapolate nonlinear optical properties. (15).

The simplified relation, expressed in terms of the linear refractive index,  $n_d$ , at the d-line of He and the Abbe number,  $\nu$  (the reciprocal of the wavelength dispersion of the linear refractive index of the material at this wavelength), is

$$n_2 = K' \frac{n_d - 1}{\nu_d^{5/4}}.$$
 (6)

This model assumes an isotropic system and a single component which determines linear and nonlinear indices. For oxide glasses, this component is oxygen.

This relation is more accurate for low-index fluoride and oxide glasses. Also, since it includes no explicit treatment of the wavelength dispersion of  $n_2$ , it is applicable only for materials and measurement wavelengths satisfying the long wavelengths approximation  $\omega << \omega_0$ . Thus it is not accurate, for example, for many chalcogenide glasses at wavelengths for which it is not possible to determine  $\nu_d$ .

Glass is an optically isotropic medium due to a lack of the long-range periodicity. On the short range scale of atomic distances, inorganic glasses are structurally ordered, but atomic sites are physically inequivalent. On the intermediate range scale of distances in the way structural units are linked together into complexes, some order may persist. The structural complexes formed in the multicomponent glasses modify and in some instances dominate the nonlinear optical properties of these glasses.

Many glass systems have been investigated by introducing a variety of modifiers into the glass network to control their nonlinearity. A systematic study of the relationship of optical nonlinearities and glass composition was conducted in the titania silicate system (16). The conclusions from this study were that the nonlinear index in this system is dominated by the nonlinear bond polarizability (hyperpolarizabilities) of the Ti-O and Nb-O bonds rather than by other structural entities such as nonbridging oxygen ions.

In general both anions and cations contribute to the bond polarizability, but for certain systems the contribution from one of the ions may dominate. For example, in oxide glasses one finds high linear and nonlinear refractive indices in systems containing large, polarizable cations. Glass in the PbO-Bi<sub>2</sub>O<sub>3</sub>-Ga<sub>2</sub>O<sub>3</sub> system developed for transmission beyond 8  $\mu$ m has a very large n<sub>2</sub> which is attributed to the presence of heavy metal cations. Hall et al. (17) obtained a correlation between  $\chi^{(3)}$  and the composition of these oxide glasses assuming that the nonlinearity is dominated by the highly polarizable lead and bismuth cations whose contributions per ion are approximately equal.

Similarly, it was found in PbO-SiO<sub>2</sub> and PbO-B<sub>2</sub>O<sub>3</sub> systems that third order nonlinear susceptibilities increase with increasing lead content (18).

However, the most significant changes in the linear and nonlinear optical properties are achieved by replacing the entire glass forming network, not just modifying it. Different oxide glasses with higher nonlinearities were examined, such as phosphate (19,20), borates (18,19) gallates (17) and tellurites (20). Tellurite glasses, for example are characterized by high linear and nonlinear index of refraction. The nonlinear index of refraction in tellurite glasses is reported to be 40-50 times that of SiO<sub>2</sub> (21,22) and it is attributed to the unique

structure of TeO2 glass.

Lines (23) had established three categories of candidate oxide glasses: the empty d-band transition metal (TM) oxides, the tellurites, and the post-TM heavy metal oxides. He calculated for each the wavelength dependence of the ratio  $n_2(\lambda)/\alpha(\lambda)$  (where  $\alpha$  is the intrinsic attenuation). From the stand standpoint of this "figure of merit" he identified the alkali metal TM gallate and TM tellurite glasses as candidates for operation throughout the wavelength range between 1 - 2.5  $\mu$ m.

#### Nonoxide Glasses

A major concern for high-power lasers is to minimize the nonlinear index so as to prevent catastrophic self focusing and beam breakup in bulk glass optical materials. Therefore, the bulk glass used for high power laser research has very low n<sub>2</sub>. The smallest linear and nonlinear refractive indices for known inorganic glass forming system occur for simple BeF<sub>2</sub> (24).

Optical nonlinearities are greater for chalcogenide glasses than for oxide or fluoride glasses. This is attributed to the replacement of the anion by sulphur or selenium, which leads to a reduction of the band gap, and the densification of the resulting glass network. The  $\chi^{(3)}$  values for various chalcogenides glasses reported by Nasu et al. (25) were measured at  $\sim 2\mu$  m to reduce the resonant contribution to the optical nonlinearity.

Lines (23) also points out that in spite of significantly enhanced  $n_2$  values, the bad news for all sulphide and selenide glasses is the fact that their Rayleigh loss coefficients are also larger than in SiO<sub>2</sub>. Therefore neither material enhances the  $n_2/\alpha$  ratio beyond that of silica at 1 - 1.5  $\mu$ m.

#### Limits of Nonresonant n<sub>2</sub>

Figure 1 compares the nonlinear index of several oxide glasses measured at  $\sim 1 \mu$  m and nonoxide glasses. As noted above, a lower limit on  $n_2$  is provided by simple BeF<sub>2</sub> glass (24). Hall et al. (17) have used an empirical approach to set upper limits for nonresonant  $\chi^{(3)}$ 

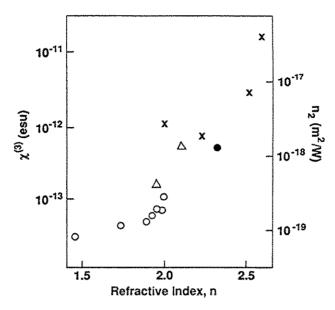


Figure 1. Nonlinear index coefficient  $(n_2)$  and third-order susceptibility  $(\chi^{(3)})$  as a function of linear index of refraction  $(n_0)$  for "x" nonoxide (chalcogenide) (Ref.25), " $\Delta$ " tellurite (Refs. 21, 23) " $\bullet$ " gallate (Ref. 17) and "o" oxide (silicate, phosphate) glasses (Refs. 1, 14).

and  $n_2$  of about  $10^{-16}$  esu or  $10^{-18}$ m<sup>2</sup>/W for oxide glasses containing heavy metal ions. The combined effect of light transition metal (Ti) and heavy metal elements (Bi,Tl) on optical properties of oxide glasses has also been addressed. Again the resulting  $n_2$  was not higher than  $10^{-18}$ m<sup>2</sup>/W (20). The results for phosphate and tellurite glasses also suggest that the practical upper limits of these glass systems is about  $10^{-18}$  m<sup>2</sup>/W. Although the  $n_2$  of oxide glasses modified by elements heavier than Bi might be larger, the practical aspects of toxicity, radioactivity, and the optical quality of actinide containing glasses have to be considered for any practical applications.

At present there is insufficient data to speculate about the upper limits of  $n_2$  for nonoxide glasses such as chalcogenides or for doped glasses. The nonlinear optical properties of these materials and their applications continue to be the subject of research effort of many groups around the world.

A global representation for n<sub>2</sub> concludes that a search for high n<sub>2</sub> glasses should pursue

those with small Sellmeier gaps (subject to the two-photon limitation), large bond length, high packing fractions and a tendency towards covalency (23).

# Metal-Doped Glasses

Glasses containing very small metallic particles can exhibit an efficient, fast optical Kerr effect (26). The large increase in refractive index resulting from implantation of 3d transition group elements in fused silica, and the interaction between implanted ions and intrinsic defects in silica have been reported (27). Ion implantation used to fabricate these composites can also cause changes in the refractive index due to compaction of the glass structure from high irradiation damage. Refractive index increases as large as 15% at 633 nm were observed for Cu (6x10<sup>16</sup>ions/cm<sup>2</sup>) using ellipsometry. These effects are due to ionic rather than metallic impurities which are thought to form a colloidal phase in the glass.

Recently the Au:SiO<sub>2</sub> composites have been shown to have nonlinear susceptibilities several hundred times larger than those of colloidal melt glasses (28).

## APPLICATIONS

The main interest in nonlinear glasses stems from their potential application in all-optical devices (29). In such devices, the optical signal affect the medium, and thereby its own propagation or the propagation of other optical signals. Nonlinear interactions can be extremely fast, and together with the picosecond and femtosecond light pulses that are routinely produced by modern laser techniques they can lead to devices that operate orders of magnitude faster than comparable electronic devices. It is this ultrafast capability that drives the quest for all-optical devices. In many of the proposed devices light propagates in fibers or waveguides. This geometry is particularly useful because in a waveguide the light propagates while confined to a small cross-sectional area; this confinement enhances the nonlinear interaction, which is proportional to the local intensity and the interaction length.

Fiber drawing and ion exchange for channel or planar waveguide fabrication has been used for many years to produce practical devices. It is natural that researchers are now looking to the same technologies for producing useful nonlinear devices. The choice of material for these devices depends on their particular design. Several material figures-of-merit have been defined, and they can be useful in choosing from a variety of comparable nonlinear materials. For example, many all-optical switching devices operate through the induction of a nonlinear phase shift on the light wave through the optical Kerr effect. A fundamental limitation for such devices is imposed by two-photon absorption, which causes nonlinear loss to the light. Since both effects (the Kerr effect and the two-photon absorption) scale the same with light intensity and interaction length, the only parameter that determines the usefulness of the system is the ratio of two nonlinear parameters, and the criterion is

$$2\beta \lambda/n_2 < 1. \tag{24}$$

This condition is violated, for example, for the lead silicate glass SF 6 at the wavelength of 532 nm, where  $n_2 = 19x10^{-20} \text{m}^2/\text{W}$  and  $\beta = 23 \text{ cm/GW}$ . However, at 1060 nm in a single mode lead silicate fiber of a similar composition, Newhouse et al. (30) report that two-photon absorption and stimulated Raman scattering did not significantly degrade the alloptical switching behaviour.

To date, the most advanced all-optical switching demonstrations have been done using standard silica optical fibers (29). Silica fibers have extremely low linear and nonlinear absorption at the wavelength of 1.5  $\mu$ m, where most interest for the telecommunication is. However, because of the small nonlinear Kerr parameter of silica, long fiber sections are required to keep the switching energy down to acceptable levels. For example, to induce nonlinear phase shifts of order  $\pi$  (as required for many switch designs) with pulses with energy of 1 pJ, a minimal fiber length of 0.5 km is required. This length is imposing many limitation on these devices. It may be advantageous to trade some of the excellent transparency for higher nonlinearity, so that the device length can be reduced to about 1 meter. Stegeman & Stolen (31) have addressed the question of the ideal material for ultrafast all-optical switching devices. For switching with one watt peak power (for example, 1 ps pulse with 1 pJ energy) in a fiber geometry, a nonlinearity 500 times that of silica is needed with attenuation up to 0.5 dB/m in a 1 meter long fiber.

In integrated optics devices, 1 cm long channel guides require a minimum nonlinearities  $n_2 = 10^{-16} \text{m}^2/\text{W}$  and a linear loss coefficient of  $0.2\text{cm}^{-1}$  for peak switching powers of 1 watt. Several techniques have been used for waveguide fabrication. Ion exchange is one of the simplest methods for changing the optical properties in the surface of the glass (32). If, however, the glass composition is not suitable for ion exchange, then thick film deposition is a possible alternative. Without a doubt, optical fibers are excellent waveguides, but integrated photonic circuits motivated the search for alternative fabrication methods of highly nonlinear optical materials. Thin films of semiconductor doped glasses (33,34) and titania silicate glasses (35) were produced by RF sputtering.

# Solitons and Spatial Solitons

Solitons are perhaps the most impressive nonlinear phenomenon to be demonstrated in optical fibers, and they are now seems to be ready for application in real communication systems. Solitons are pulses of a specific shape, power, and width that propagate along a nonlinear transmission medium without changing their shape (36). Soliton propagation has been demonstrated in silica based optical fibers in the anomalous dispersion regime, i.e. at wavelengths longer than 1.3 µm. The intensity-induced refractive index changes can then compensate for the dispersion, leading to a pulse propagating over large distances without broadening. Solitons are considered for ultra-long distance communications, such as in trans-oceanic cables. Soliton transmission over 10,000 km has been demonstrated in dispersion-shifted fiber (37).

Solitons have been called 'natural bits' - they tend to keep their pulse shape without smearing or broadening. They are therefore very attractive for communications application, and for this reason they have also been considered for logic and switching devices (29,38,39). As mentioned above, for such devices it will be useful to have fibers about 500 times more nonlinear than silica. It is important to remember that any all-optical device in fibers that is designed to operate with short pulses must meet requirements not only on nonlinear parameters but also on dispersion. For soliton devices in particular, these fibers must have anomalous dispersion at the wavelength of operation in order to compatible with

soliton propagation.

In analogy to temporal solitons, spatial solitons are beams that propagate in a nonlinear medium without changing their shape. In this case self-focusing due to nonlinear index changes compensates for diffraction (40,41). The diffraction is changing in the spatial domain, therefore the name spatial soliton. These self-trapped beams are known to be unstable in a bulk optical medium where the self-focusing effects lead to a collapse of the beam to a very tight spot and usually to catastrophic damage. If, however, the propagation of light is limited to a two-dimensional medium such as in a planar waveguide structure, a stable self-induced waveguide can be formed. Spatial solitons have also been proposed as building blocks for integrated all-optical switching circuits and devices (42).

#### CONCLUDING REMARKS

Nonlinear optical phenomena have now been investigated from the infrared to the ultraviolet region in a wide range of glass compositions. Driven by applications requiring either very low or very high nonlinear refractive indices, almost the entire periodic table of elements has been exploited in tailoring the optical Kerr effect in known glass forming systems. Data on the wavelength dispersion of the nonlinear response of glass is sparse, however. More systematic investigations of  $n_2$  and  $\beta$  in the resonant regime are needed to complete our understanding and treatment of optical nonlinearities. This is particularly important for materials optimization in applications where a tradeoff may be necessary between large  $n_2$  to reduce power and small  $\beta$  to reduce losses.

Whereas intrinsic effects in known inorganic glass systems have been widely explored, the nonlinear behaviour associated with extrinsic effects of some dopants has been investigated only recently. Many more possibilities with respect to dopants, hosts, and processing remain to be discovered and fully characterized.

The main drive to study nonlinear glasses comes from their potential application in ultrafast all-optical switching devices. All-optical device are studied in many laboratories around the world, and many groups have demonstrated various principles that can be used to implement them. However, at this time, all-optical devices are still a lab curiosity. Materials issues are probably the most important factors limiting the successful implementation of all-optical devices. The nonlinear properties are only one of many considerations that usually enter into the final selection of materials, therefore the role of glass in many applications of nonlinear phenomena remains to be established. However, because of its extensive compositional versatility and its demonstrated availability in varied physical forms and dimensions, glass presents many attractive scientific and technological possibilities.

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#### Profile

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