Inorganic Glasses with Optical Nonlinearity

Hiroyuki Nasu

Department of Chemistry for Materials, Faculty of Engineering, Mie University, Kamihama, Tsu 514, Japan

The second- and third-order optical nonlinearity of Inorganic homogeneous glasses have been extensively studied recently. This paper reports the second harmonic genaration from OH containing silica glasses, and the third harmonic generation from various glasses. Second harmonic generation increases with increasing OH concentration. The third harmonic generation intensity is basically dependent on the refractive index of glasses.

1. Introduction

Recently, optical nonlinearity has extensively studied from not only academic interests but practical use for photonic signal transmitting and processing systems. Thus, many works have been carried out for developing materials with large optical nonlinearity. The polarization formed (P) from the incident light beam intensity (E) can be expressed as the following equation.

$$P=\chi^{(1)}E+\chi^{(2)}E\cdot E+\chi^{(3)}E\cdot E\cdot E+\cdots\cdot(1),$$

where $\chi^{(1)}$ is linear optical susceptibility and $\chi^{(2)}$ and $\chi^{(3)}$ are the second- and third-order optical susceptibility, respectively. $\chi^{(2)}$ should not theoretically appear from the materials with optical inversion symmetry, and thus it had been believed glasses should not have $\chi^{(2)}$. However, The second harmonic generation (SHG) was measured from Ge-doped SiO_2 glass fiber[1] and also from thermally poled SiO_2 glass[2]. Based on these works, we tried thermal poling for various glasses.

On the other hand, $\chi^{(3)}$ can be seen in all substances. To develop large $\chi^{(3)}$ with fast response and relaxation time, nonlinear polarization can be considered as quite useful effect. Therefore, some homogeneous glasses has been prepared and measured their $\chi^{(3)}$. In this paper, the nonlinear optics involving both $\chi^{(2)}$ and $\chi^{(3)}$ of homogeneous glasses are explored.

2.Experimental Procedure

2.1 $\chi^{(2)}$ in the silica glass

Second-harmonic genaration (SHG) caused by the second-order optical nonlinaerity is convinient to probe the existence of the second-order optical nonlinearity in the materials. The samples were SiO_2 glasses prepared by various technique such as VAD (Vapor Axial Deposition), conventional melting and quencing and sol-gel method. The samples were polished and Au electrodes were deposited on the both side of the samples. Then, dc electric field was applied at various temperature for various time duration, and cooled down to the room temparature with applying electric field. Subsequentry, electrodes was eliminated. SHG measurements were carried out at a pump wavelength of 1.06μm using a Q-switched Nd:YAG laser with a pump pulse duration of 5.5ns and repetition rate of 10Hz. In this experiment, the d_{11} generation of quartz was used as reference.

2.2 $\chi^{(3)}$ of the glasses

$\chi^{(3)}$ of homogeneous glasses was evaluated by the third-harmonic generation (THG). Various homogeneous glasses were used for THG measurement. Those were typical chalcogenide (for instance, As_2S_3 and GeS_3), tellurite ($PbO-TiO_2-TeO_2$, $Li_2O-TiO_2-TeO_2$), germanate (Na_2O-GeO_2), silicate (R_2O-SiO_2; Na_2O-SiO_2, Li_2O-SiO_2, K_2O-SiO_2, Rb_2O-SiO_2, Cs_2O-SiO_2) and CdO-containing glasses. They were pre-pared by the conventional melting and quenching technique. The melts were quenched to room temperature and the surfaces were polished to eliminate surface light scattering.

The THG values of the glasses were measured using a pump wavelength of 1.9μm generated from H_2 Raman shifter exited by a Q-switched Nd:YAG laser. The pump duration was 5.5ns, the repetition rate was 10Hz and the peak power density was $200MW/cm^2$.

3.Results and Discussion

3.1 $\chi^{(2)}$ of the glasses

Fig.1 shows a typical fringe pattern. one can see clear oscillation pattern which can not found in the data in ref.2). The calculated poled layer was nearly 1mm and thus almost whole body is polarized. The dependence of SHG intensity on OH concentration are shown in Fig.2. One can see the increase of OH concentration increases SHG intensity, and the largest intensity is seen from sol-gel derived SiO_2 glass with the largest OH concentration. The poling condition also influences the intensity. The poling duration, temperature and dc voltage were varied. SHG was observable for the glasses poled over 150°C and over 1kV. The largest SHG was seen by the poling at 200°C nd 3kV, the derived $\chi^{(2)}$ corresponds to 8% of that of d_{22} of $LiNbO_3$.

3.2 $\chi^{(3)}$ of the glasses

There is semiemprical rule between $\chi^{(1)}$ and $\chi^{(3)}$, which is called Miller rule as

$$\chi^{(3)}=\{\chi^{(1)}\}^4 x10^{-10}(esu) \qquad (2)$$

$$\chi^{(1)}=(n^2-1)/4\pi \qquad (3)$$

where n is the refractive index. Thus, the glasses with high refractive index should have large $\chi^{(3)}$. The relationship between $\chi^{(3)}$ and n is shown in Fig.3. Basically, it can be said that high-index glasses have large $\chi^{(3)}$. The largest $\chi^{(3)}$ is so far found from chalco-

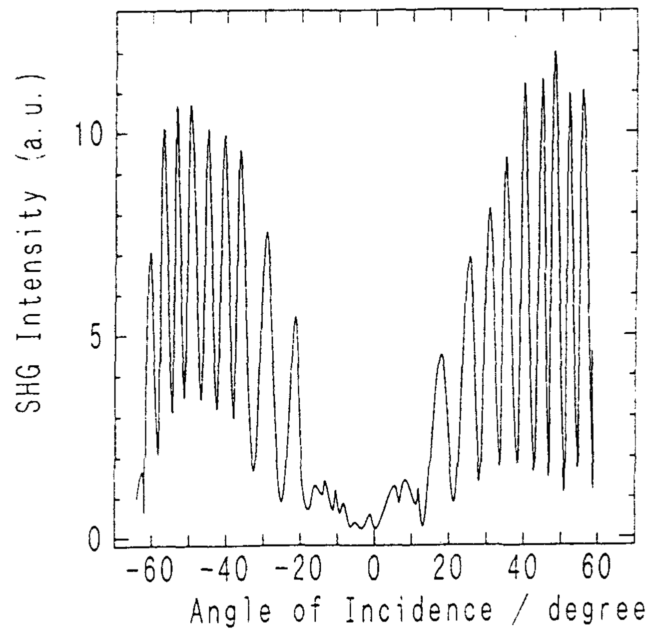

Fig.1 Typical fringe pattern of SHG

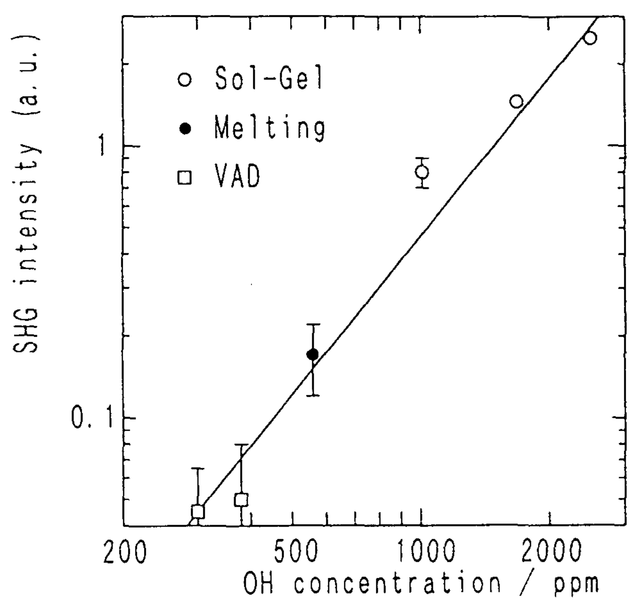

Fig.2 SHG intensity as a function of OH concentration.

genide glasses in the order of 10^{-11}–10^{-12} esu. Among the oxide glasses, TeO_2-based glasses have large $\chi^{(3)}$. The contribution of each structure unit can be analyzed by the estimation of hyperpolarizability ($\alpha^{(3)}$) for each unit. From the estimation, large ions, dense structure and nonbridging ions were found to be effective to enlarge $\chi^{(3)}$.

4. Conclusion

$\chi^{(2)}$ and $\chi^{(3)}$ of homogeneous glasses were explored from SHG and THG, respectively. SHG relates to the concentration of OH, and also depends on the poling condition. The largest $\chi^{(2)}$ can be observable by the poling at 200°C. From the oscillation pattern, the almost whole body was poled. On the other hand, THG intensity basically depends on the refractive index of the glasses. The estimation of hyperpolarizability of each structure unit revealed large ions, dense structure, and nonbridging ions enlarge $\chi^{(3)}$.

References

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2) R.A. Myers, N. Mukherjee and S.R.J. Brueck, Opt. Lett. 16 (1991) 1732.

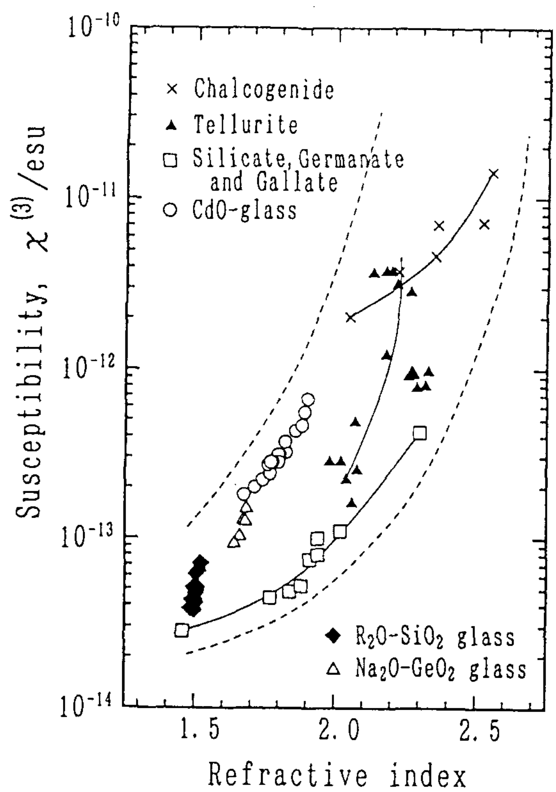

Fig. 3 $\chi^{(3)}$ as a function of refractive index.