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Third-order nonlinear optical properties of chalcogenide glasses

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Third-order nonlinear optical properties of chalcogenide glasses were investigated through third-harmonic generation (THG), optical Kerr shutter (OKS), and degenerate four-wave mixing (DFWM) measurements. We examined the dependence of the THG susceptibility on the absorption edge, thereby showing that the susceptibility rapidly increased as the absorption edge red shifted. Moreover, the THG susceptibility was compared with the OKS susceptibility. It was found that both susceptibilities almost coincided within the range of experimental errors. The OKS and DFWM experiments indicated that a high-speed compact optical switching device was obtained using the chalcogenide glasses. © 1997 American Institute of Physics. [S0003-6951(97)02508-4]

Ultrafast optical switching operation is considered to be an important feature in applications to the future high-bit-rate handling of optical signals. Nonlinear optical glasses have been developed as promising candidates for constructing these optical signal operation systems, accentuating many advantages such as large nonlinearities, excellent processability, and high transparency. 1–5 For instance, a chalcogenide glass has demonstrated highly efficient optical Kerr shutter (OKS) switching operation with picosecond response time. 5 However, the optical switch using the chalcogenide glass required a meter-sized fiber and larger nonlinearity has been needed for making a compact switching device. Furthermore, few switching demonstrations have been done in the subpicosecond level. In this letter we investigated the third-order nonlinear optical properties of the chalcogenide glass in order to yield a larger nonlinear material with ultrafast response time. Third-harmonic generation (THG) measurement and OKS experiment were conducted to bring out larger nonlinearity in the chalcogenide glass. The response time was also examined by means of degenerate four-wave mixing (DFWM) technique.

The THG measurement was carried out using the pump light with the wavelengths from 1.9 to 2.2 μm. The pump pulse duration was 6 ns and the repetition rate was 10 Hz. The third-harmonic (TH) wave was detected with a photomultiplier tube and a boxcar averager. The pump power density varied from 50 to 100 MW/cm². The THG susceptibility \(\chi^{(3)}\) (THG) value was determined by comparing TH intensity with the standard of fused silica glass, as shown by the following equation: 6

\[
\chi^{(3)}(\text{THG}) = \left( \frac{n+1}{n_S+1} \right)^4 \frac{I_{3s}}{I_c} \left( \frac{I_{3s}}{I_{3w_S}} \right)^{1/2} \chi^{(3)}(\text{THG}).
\]

Here \(n\) is the refractive index, \(I_c\) is the coherence length, and \(I_{3w}\) is the TH intensity. The suffix \(S\) means the standard medium. We used the \(\chi^{(3)}(\text{THG})\) value reported by Meredith, Buchalter, and Hanzlik. 7

In the OKS experiment, a Nd:YAG nanosecond laser (1.064 μm wavelength; 6 ns duration; 10 Hz repetition) and a Nd:YAG laser-pumped nanosecond dye laser (0.70 μm wavelength; 6 ns duration; 10 Hz repetition) were used to generate a gate beam. A laser diode pulse with a wavelength of 0.81 μm was used as a probe beam. The duration was 50 ns. The photomultiplier tube was employed as a detector. The polarization of the gate beam was set at π/4 with respect to that of the probe beam. The gate power density was around 50 MW/cm². We calculated the OKS susceptibility \(\chi^{(3)}(\text{OKS})\) by comparing the probe transmittance intensity with the standard of carbon disulfide under the same gate power. 8,9 The \(\chi^{(3)}(\text{OKS})\) value is given by

\[
\chi^{(3)}(\text{OKS}) = \left( \frac{n}{n_S} \right)^2 \left( \frac{T}{T_S} \right)^{1/2} \chi^{(3)}(\text{OKS}),
\]

where \(n\) is the refractive index, \(T\) is the probe transmittance, and the suffix \(S\) indicates the standard medium.

The boxcar’s configuration was made when conducting the DFWM measurement. The beam wavelength of 0.81 μm was produced by the combination of a Ti:Al₂O₃ regenerative amplifier system and an optical parametric amplifier. The pulse duration was 200 fs and the repetition rate was 200 kHz. The pump power density was around 50 MW/cm². The photomultiplier tube was used to detect the DFWM signal. We delayed the probe beam against the two pump beams for investigating the response time.

Figure 1 shows the dependence of the measured \(\chi^{(3)}\) (THG) value on the absorption edge of the chalcogenide glass. The chalcogenide glass provided various absorption edges by changing the component atoms. Both THG susceptibility and absorption edge were estimated by using a 0.5-mm-thick medium. Since the chalcogenide glass had wide-ranged densities, i.e., from 2.5 to 3.3, all the \(\chi^{(3)}(\text{THG})\) values were calculated for 1 M concentration to evaluate the nonlinearity of the same concentration. In Fig. 1, the 1 M \(\chi^{(3)}(\text{THG})\) value exponentially increased as the absorption edge red shifted and, in a closer view, the exponential slope was found to be approximately six, which sufficiently coincided with the prediction by a theoretical work. 10 Larger
nonlinearity was obtained by using As and Se atoms instead of Ge and S atoms, respectively. The maximum $\chi^{(3)}$ (THG) value as a bulk reached $1.4 \times 10^{-11}$ esu in the case of As$_{40}$S$_{57}$Se$_3$. For the chalcogenide glass, increasing Se atom fraction is one of the effective keys to yielding larger nonlinearity.

The OKS property of the As$_{40}$S$_{57}$Se$_3$ bulk which exhibits the largest THG susceptibility is shown in Fig. 2. The transmittance of the probe light was plotted as a function of the gate power. The result of carbon disulfide was also shown in Fig. 2 for comparison. The As$_{40}$S$_{57}$Se$_3$ bulk had a thickness of 3 mm. The probe transmittance of the OKS measurement is given by

$$T \propto \sin^2[\chi^{(3)}(OKS) P_g^i]$$

where $P_g$ is the gate power. The probe transmittance of the As$_{40}$S$_{57}$Se$_3$ bulk was observed to follow Eq. (3), thus confirming the OKS operation. It was also found that the probe transmittance of the As$_{40}$S$_{57}$Se$_3$ bulk was almost twice as large as that of carbon disulfide. The influence of two-photon absorption (TPA) was not seen in this experiment. The chalcogenide glass having a more red-shifted absorption edge reasonably reduces the gate power, which provides a compact-sized optical switching device.

In the OKS configuration, the OKS susceptibility $\chi^{(3)}(OKS)$ is written as

$$\chi^{(3)}(OKS) = \chi^{(3)}(OKS) - \chi^{(3)}(SPM).$$

Here the designations $\parallel$ and $\perp$ refer to the parallel and perpendicular directions relative to a linearly polarized gate light. The $\chi^{(3)}(OKS)$ is equal to $(1/3) \chi^{(3)}(OKS)$ for the case of solid media and the OKS susceptibility tensors are considered to be twice those values defined in self-phase modulation (SPM), so we can obtain the following relationship:

$$\chi^{(3)}(OKS) = t \chi^{(3)}(SPM),$$

where the degeneracy factor is included in the third-order susceptibility. Note that this is not the Maker–Terhune definition for the third-order susceptibility. Similarly, the nonlinearity of THG can be related to that of SPM, by supposing that the THG susceptibility tensors are one-third of the SPM ones. That is,

$$\chi^{(3)}(THG) = \frac{1}{3} \chi^{(3)}(THG).$$

Using Eqs. (5) and (6), it is indicated that the OKS and THG susceptibilities are both represented by the same SPM susceptibility, and because of this we can conclude that the OKS susceptibilities are known by the data of the simple THG measurement. Table I supports this conclusion experimentally. Here, we listed the measured susceptibility ratios of OKS to THG, $\chi^{(3)}(OKS)/\chi^{(3)}(THG)$. The ratios were found to almost coincide within the range of experimental errors. Table I also shows that all of these ratios take the values of less than 0.3, although the SPM susceptibility for the OKS measurement should have the equal value with that for the THG measurement. This might, like in the case of organic low-molecular-weight compounds, favor the previous works pointing out that the nonlinearity of silica glass has been overestimated to be two or three times larger than the actual value.

The response behavior of the chalcogenide glass bulk is shown in Fig. 3. We used a 0.5-mm-thick As$_{20}$S$_{80}$ bulk for the DFWM investigation. The DFWM intensity was plotted as a function of the delay time. The As$_{20}$S$_{80}$ bulk was found to have the response time as fast as the pump pulse. The signal increased in proportion to the cubic of the pump beam intensity, and any slow decay was not observed with the As$_{20}$S$_{80}$ bulk. Hence, the electric polarization effect, whose relaxation time is less than subpicosecond, is plausibly the main third-order nonlinear mechanism of the chalcogenide glass. Thermal effect or TPA effect was not considered the origin of the third-order nonlinearity. If we employ a faster pump pulse, the chalcogenide glass can achieve high-speed switching operation.

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In conclusion, the THG, OKS, and DFWM measurements clarified the third-order nonlinear properties of the chalcogenide glass. It was shown that the THG susceptibility increased in proportion to the sixth power of the absorption edge, and that the susceptibility ratios of OKS to THG had the almost constant values of less than 0.3. The OKS measurement exhibited that the As$_{40}$S$_{80}$Se$_3$ chalcogenide glass provided twice probe transmittance of the conventional carbon disulfide. The DFWM experiment also pointed out that an ultrafast response time of less than subpicosecond was attainable with the chalcogenide glass. The chalcogenide glass will produce a compact optical switching device having high-speed operation time.

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