Nonlinear optical characterization of phosphate glasses based on ZnO using the Z-scan technique*

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The nonlinear optical properties of a phosphate vitreous system $[(ZnO)_x-(MgO)_{30-x}-(P_2O_5)_{70}]$, where $x = 8$, 10, 15, 18, and 20 mol% synthesized through the melt-quenching technique have been investigated by using the Z-scan technique. In the experiment, a continuous-wave laser with a wavelength of 405 nm was utilized to determine the sign and value of the nonlinear refractive (NLR) index and the absorption coefficient with closed and opened apertures of the Z-scan setup. The NLR index was found to increase with the ZnO concentration in the glass samples by an order of 10^{-10} cm^2·W^{-1}. The real and imaginary parts of the third-order nonlinear susceptibility were calculated by referring to the NLR index (n_2) and absorption coefficient (β) of the samples. The value of the third-order nonlinear susceptibility was presented by nonlinear refractive or absorptive behavior of phosphate glasses for proper utilization in nonlinear optical devices. Based on the measurement, the positive sign of the NLR index shows a self-focusing phenomenon. The figures of merit for each sample were calculated to judge the potential of phosphate glasses for application in optical switching.

Keywords: Z-scan technique, nonlinear refractive index, nonlinear absorption coefficient, phosphate glasses

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1. Introduction

The examination of nonlinear materials such as glasses has a significant function in determining their potential for use in optical devices.[1,2] Large nonlinear and linear refractive indices and small absorption coefficient warrant research interest in many fields such as optics and photonics.[3,4]

Glasses have currently been used in a broad range of optical applications because changing their composition can make them appropriate for use in nonlinear optical fields.[5] Glass is the most important and the newest material that can be applied in many fields such as optics and photonics.[6,7] Among different glasses, phosphate glasses possess a diversity of scientific and technological implementations because of their suitable optical and electrical properties.[8] ZnO is a semiconductor with large band gap (band gap energy: 3.4 eV, excitation binding energy: 60 meV) which has been the focus of numerous scientific studies due to its applications in different devices such as laser diodes.[9]

The Z-scan method is one of the simplest and most precise methods for characterizing nonlinear materials by using nonlinear absorptive (via open aperture) and nonlinear refractive (NLR) (via closed aperture) behavior.[10,11] In the Z-scan technique, the third-order nonlinear susceptibility as a nonlinear response of medium to the field is used by applying both the NLR index and the absorption coefficient.[12] The single-beam Z-scan technique was used to scan the sample in the far field through an aperture on a focal plane based on the Gaussian beam decomposition method.[13–15] In this method, the sample is scanned using a focused laser beam on a focal plane.

When a dielectric material enters the electromagnetic field, it stimulates electric polarization.[16] In this material, influence of the electric field causes a deformity in the spatial distribution between the electrons and the nucleus of atoms.[16] The distortion leads to electric dipoles and can be manifested as polarization.[16] When the intensity of the electric field is low, the induced polarization is only proportional to the electric field However when the intensity of the electric field is high, the polarization is not field-dependent and the susceptibility becomes field-dependent.[16,17] Therefore, polarization (P) turns into a nonlinear function of high-power electric fields: $P = \varepsilon_0(\chi^{(1)}E + \chi^{(2)}E.E + \chi^{(3)}E.E.E + \cdots)$.[17,18] In this study, ε_0 is the permittivity of vacuum and $\chi^{(3)}$ is the third-order nonlinear susceptibility consisting of two parts, namely, real and imaginary, as signified by the NLR index and the absorption coefficient respectively.[19] The figure of merit $(F < 1)$ for the optical nonlinear media is specified to make them appropriate for all optical switching applications.[20,21] Reported results of nonlinear characterization were demonstrated by applying the pulse laser at different wavelengths.[22–24] In this work, a continuous wave (CW) with wavelength of 405 nm was utilized to measure the nonlinear properties. This study demonstrates the effect of ZnO on the optical properties of the phosphate glassy structure to show

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their potential for application in optical devices.

2. Experiment

Melt-quenching technique was applied to prepare the phosphate glassy system $[(ZnO)_x-(MgO)_{30-x}-(P_2O_5)_{70}]$ ($x=8$, 10, 15, 18, and 20 mol%). The materials were weighed and mixed. Afterward, they were transferred in an alumina crucible and melted. They were casted and annealed in a furnace. After cooling, the samples were polished and cut.[25]

As illustrated in Fig. 2, the samples were characterized using a Z-scan setup to determine the NLR index and the nonlinear absorption coefficient in closed- and open-aperture setups respectively. In this work, the ternary zinc–magnesium phosphate glass sample in the translation stage of the Z-scan setup was scanned using a CW laser at 405 nm. The lens focused the laser beam on a small dot to characterize the samples in the direction toward the z axis. The focused laser light which has passed through the sample was detected by a detector at room temperature. The output power of laser was approximately 36 mW at the focus point.

3. Results and discussion

As shown in Fig. 1, the absorption spectra of the phosphate glass samples were measured in the range from 190 nm to 600 nm. The ultraviolet–visible (UV–Vis) absorption spectra indicate typical absorption plots for the two glassy samples $[(ZnO)_8-(MgO)_{22}-(P_2O_5)_{70}]$ and $[(ZnO)_{18}-(MgO)_{12}-(P_2O_5)_{70}]$. The absorption peak of 8 mol% ZnO (235 nm) was shifted to a higher wavelength (240 nm) where the ZnO concentration was 18 mol%.

To measure the NLR index, the transmitted light was used to scan the sample through the Gaussian beam waist in the direction of the z axis where the transmitted light is a function of z.[26] In the normalized transmittance curve, the peak and valley positions are related to the sign of the phase shift ($\Delta\varphi$) which is related to the z axis.[27] The negative sign of the phase shift of the sample reveals a self-defocusing phenomenon unless it is a self-focusing phenomenon correlated with the sample properties.[28–30] Given that the sample is far from the focal area of the focusing lens, the light intensity is weak and the sample refraction remains unchanged Therefore nonlinear behavior becomes insignificant.[31] For the self-defocusing effect, as the sample drew closer to the focal area, the incident light was collimated and caused an increase in the irradiance before it reached the focal point. Following the focal point, the laser beam diverged thus reducing the irradiance.[32] The opposite behavior is called the self-focusing effect.

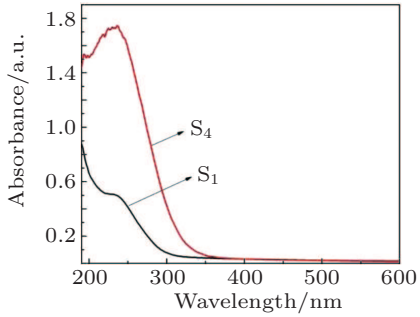


Fig. 1. (color online) UV–Vis absorption spectra of S_1 and S_4.

The NLR index is obtained by the proper fitting curve of the Z-scan equation 1.[33,34]

$$T_{CA}(z)=1-(4\Delta\varphi_0)\left[(x^2+1)(x^2+9)\right]^{-1}x, \tag{1}$$

where $\Delta\varphi_0=(2\pi n_2 I_0 L_{eff})/\lambda$ is the on-axis nonlinear phase shift of the light-induced area at the focus. I_0 is the beam intensity, $L_{eff}=(1-\exp(-\alpha L))/\alpha$ is the sample effective thickness, λ is the laser wavelength and $x=z/z_0$ is the sample position with respect to the focal plane.[35]

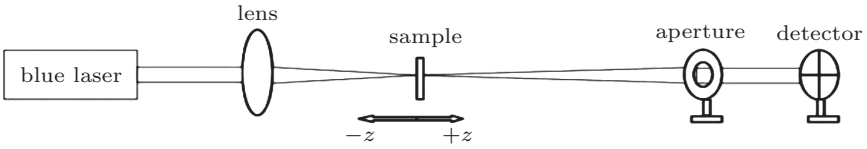


Fig. 2. Z-scan setup schematic diagram.

The nonlinear absorption coefficient was calculated using the open aperture or without aperture ($S=1$). To measure this parameter, the aperture is removed or opened completely. In this case, the absorption is the irradiance nonlinear function according to the configuration of nonlinear absorption. The function can be presented by two concepts, namely, saturation and reverse saturation absorption.[36] When the valley appears at the focus point, reverse saturation absorption occurs, the transmittance can be decreased by increasing the irradiance of input power. Meanwhile, the nonlinear absorption shows peak at the focus when it increases by increasing the irradiance input power. This phenomenon is called saturation absorption.[36] The nonlinear absorption magnitude is calculated by fitting the empirical data[37] with the following equation:

$$T(z,S=1)=\sum_{m=0}^{\infty}[-q_0(z,0)]^m(m+1)^{-3/2}. \tag{2}$$

For $|q_0(z)|<1$, $q_0(z)=\beta I_0 L_{eff}(1+z^2z_0^{-2})^{-1}$ and m is an integer.[38]

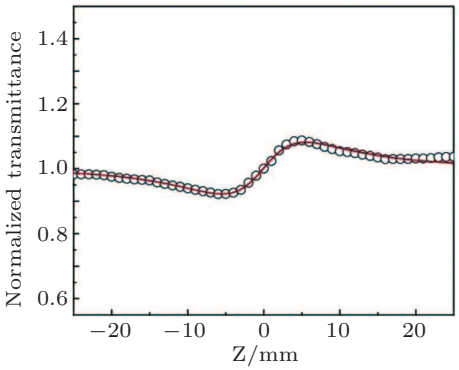


Fig. 3. (color online) Normalized transmittance curve of the closed aperture for S_1.

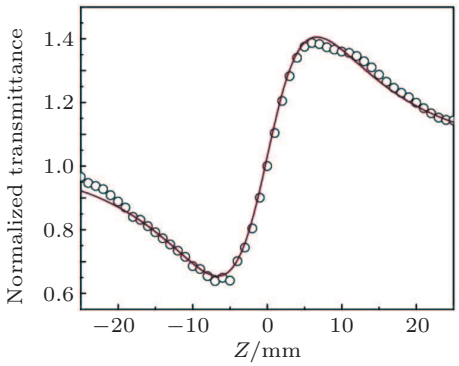


Fig. 4. (color online) Normalized transmittance curve of the closed aperture for S_4.

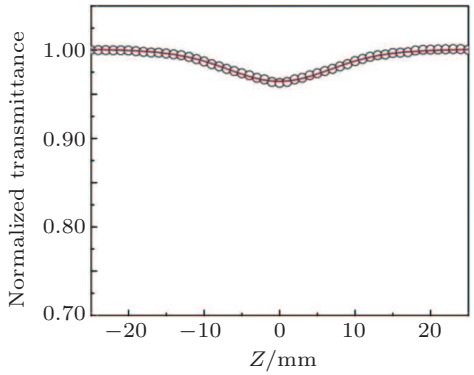


Fig. 5. (color online) Normalized transmittance curve of the open aperture for S_1.

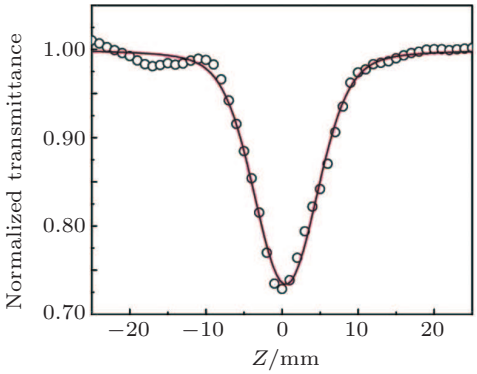


Fig. 6. (color online) Normalized transmittance curve of the open aperture for S_4.

Table 1. Nonlinear optical parameters of phosphate glasses.

Sample code	Sample	$n_2/10^{-10}$ cm^2·W^{-1}	$\beta/10^{-8}$ cm·W^{-1}	$F/10^{-1}$
S_1	$[(ZnO)_8$–$(MgO)_{22}$–$(P_2O_5)_{70}]$	0.45	0.60	0.054
S_2	$[(ZnO)_{10}$–$(MgO)_{20}$–$(P_2O_5)_{70}]$	0.49	9.68	0.800
S_3	$[(ZnO)_{15}$–$(MgO)_{15}$–$(P_2O_5)_{70}]$	1.22	4.03	0.133
S_4	$[(ZnO)_{18}$–$(MgO)_{12}$–$(P_2O_5)_{70}]$	1.56	1.34	0.034
S_5	$[(ZnO)_{20}$–$(MgO)_{10}$–$(P_2O_5)_{70}]$	2.11	5.81	0.111

The increase in ZnO mol% results in the increase in the value of the NLR index of the phosphate glasses. This finding indicates that the light energy is absorbed further as the number of ZnO molecules increases, thereby increasing the photon energy and enhancing the effect of nonlinearity. The Z-scan data obtained via closed aperture purely reflect the effects of nonlinear refraction. As indicated in Fig. 7, the NLR index as a function of ZnO shows a nonlinear behavior. The positive sign of the NLR indices (n_2) of the sample signifies a self-focusing phenomenon. The nonlinear absorption coefficients of $[(ZnO)_x - (MgO)_{30-x} - (P_2O_5)_{70}]$ $(x = 8, 18)$ glassy system calculated using a laser at a wavelength of 405 nm in the open aperture position $(S = 1)$ by the order of 10^{-8} (cm·W^{-1}) are presented in Figs. 5 and 6. The figure of merit $F = \lambda n_2^{-1}\beta$ was calculated for the phosphate samples, where λ is the wavelength and β and n_2 are the nonlinear absorption coefficient and the NLR index respectively $F < 1$ is the standard value of a suitable component in the field of nonlinear optics.[17,20,21] The sample figure of merit value shows the potential of phosphate glass to use in optical switches by following the condition of $F < 1$. The magnitudes of the NLR index (n_2), nonlinear absorption coefficient (β), and figure of merit (F) are listed in Table 1.

The third-order nonlinear susceptibility $\chi^{(3)}$ was obtained by applying the n_2 and β values in the following formulas:[39]

$$\mathrm{Re}\left(\chi^{(3)}\right) = 10^{-4}(\varepsilon_0 n_0^2 c^2/\pi)n_2, \tag{3}$$

$$\mathrm{Im}\left(\chi^{(3)}\right) = 10^{-2}(\varepsilon_0 n_0^2 c^2 \lambda/4\pi^2)\beta, \tag{4}$$

where n_0 is the linear index of refraction, c is the speed of light and ε_0 is the vacuum permittivity[19] The third-order nonlinear susceptibility $\chi^{(3)}$ contains real and imaginary parts. The real part of this complex was obtained by using the NLR index (Eq. 3) and the imaginary part was obtained using the nonlinear absorption (Eq. 4).[19]

The values of the real and imaginary parts of the nonlinear susceptibility were presented by alluding the amount of NLR index (nonlinear optical refractive behavior) and absorption coefficient (nonlinear optical absorptive behavior) as shown in Table 2.

Table 2. Real and imaginary parts of the third-order nonlinear susceptibility.

Sample code	Sample	$\mathrm{Re}(\chi^{(3)})/10^{-15}$	$\mathrm{Im}(\chi^{(3)})/10^{-5}$
S1	$[(ZnO)_8$–$(MgO)_{22}$–$(P_2O_5)_{70}]$	1.19	0.11
S_2	$[(ZnO)_{10}$–$(MgO)_{20}$–$(P_2O_5)_{70}]$	1.38	1.82
S_3	$[(ZnO)_{15}$–$(MgO)_{15}$–$(P_2O_5)_{70}]$	8.80	0.76
S_4	$[(ZnO)_{18}$–$(MgO)_{12}$–$(P_2O_5)_{70}]$	14.42	0.25
S_5	$[(ZnO)_{20}$–$(MgO)_{10}$–$(P_2O_5)_{70}]$	26.65	1.12

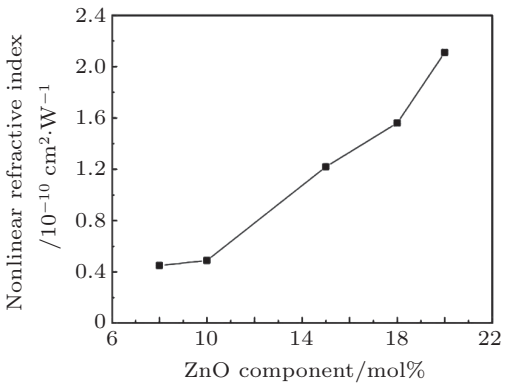


Fig. 7. NLR index of phosphate glasses as a function of ZnO component.

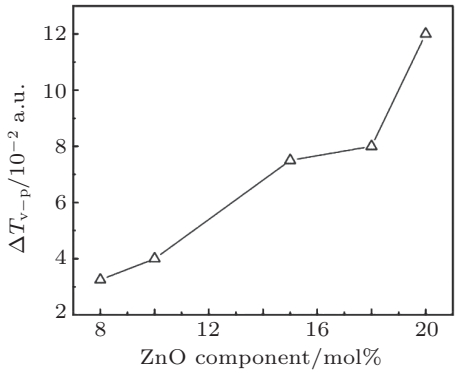


Fig. 8. Variation of $\Delta T_{\mathrm{v-p}}$ as a function of ZnO mol%.

The dependence of $\Delta T_{\mathrm{v-p}}$ (difference between valley and peak in the curve of normalized transmittance) on the ZnO component is shown in Fig. 8. Obviously, $\Delta T_{\mathrm{v-p}}$ increases with increasing ZnO component in the phosphate glasses.

4. Conclusions

The nonlinear optical properties of phosphate glasses were successfully examined using a single-beam *Z*-scan technique. By fitting the experimental data, passive parameters such as the NLR index and the absorption coefficient of phosphate glasses were determined. In this series, an increasing trend was found in the amount of NLR index by increasing the ZnO concentration. The magnitudes of NLR index and absorption coefficient were also found to appraise the third-order nonlinear susceptibility. The value of the figure of merit for each sample reveals the capability of phosphate glasses to be employed in telecommunication. All the experimental results show that the ternary zinc–magnesium phosphate glasses system is a competitive component for implementation in optical switching.

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