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Near UV Second Harmonic Generation using Anomalous Dispersion of Poled Polymer

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Second harmonic generation (SHG) by guided phase matching using anomalous dispersion of poled polymer is investigated in waveguide structure. The second harmonic TM_0 guided mode can be generated from the fundamental TM_0 guided mode and then the second harmonic power was higher than any other phase matchable mode because the overlap integral between the fundamental and the second harmonic waves is the largest in theoretical analysis. Near UV SHG (370nm) was observed from the fundamental wavelength of Ti-sapphire laser (740nm).

Keywords: second harmonic generation (SHG); poled polymer; anomalous dispersion; phase matching; guided mode; overlap integral

INTRODUCTION

Second harmonic generation (SHG) by second order nonlinear optical phenomena has been much attractive because it can be used as a shorter wavelength light source with small size for high density optical memory storage. Second harmonic generation using inorganic and organic materials have been reported[1–8]. Among these materials, organic materials have been widely investigated because of high nonlinearity, fast response time, structural flexibility, and high damage threshold[9].

For efficient SHG waveguide structure was widely used as fundamental beam can be confined over long interaction length. Also phase matching between the

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fundamental wave and the second harmonic wave is required. Three methods have been utilized for phase matching; (1) guided mode phase matching, (2) Cerenkov type phase matching, (3) quasi-phase matching.

Poled polymer, disperse red 1 (4-[N-ethyl-N-(2-hydroxyethyl)]amino-4′-nitroazobenzene) side chain copolymer with polymethyl methacrylate (PMMA), has absorption peak near 480nm and transparency window near UV range. The green and blue second harmonic generation were reported in elsewhere[10] using this poled polymer. Since the poled polymer has absorption in the green and blue wavelength they were generated from Cerenkov type phase matching.

In this paper, to generate very short wavelength using this poled polymer was studied in the waveguide structure. Guided mode phase matching can be achieved by using anomalous dispersion of poled polymer from red light to UV. And then the lowest order guided mode of second harmonic was generated from the lowest order guided mode of fundamental wave in waveguide structure. The waveguide film thickness for phase matching was decided by theoretical analysis and was good agreement with experiment. Near UV SHG was observed in poled polymer.

SAMPLE FABRICATION

Side chain poled polymer, methylmethacrylate (MMA) and DR1-substituted methacrylate (DR1MA) (DR1 10wt.%), was prepared and spin coated on a pyrex glass substrate. The pyrex glass substrate was put on the edge of the sample holder of the spinner. The fabricated waveguide layer thickness was continuously varied along the sample length because the tapered waveguide structure was required. The range of the waveguide thickness was from 1.06 μm to 1.14μm on the sample length 1 cm. It was measured by m-line method using prism coupler. Fig. 1 shows the structure of poled polymer. Ethylene glycol monoethyl ether acetate was used as the solvent of the polymer. The sample was prebaked at 80°C to remove the residual solvent and moisture, and successively corona poling process was carried out. The condition of poling process was 5kV, 15 minutes at 110 °C. The sample was placed on a grounded planar copper electrode. A 40μm-diameter tungsten wire was positioned above the sample as a positive electrode apart from the grounded electrode at 8mm distance. The sample was raised to glass temperature (T_g) of the polymer. Then the voltage was applied and the polymer film was cooled down to room temperature in the presence of the field. In the corona poling process, raising and cooling rate of temperature was controlled carefully. The absorption spectrum of poled polymer before and after poling was different as shown in previous paper[10]. The cutoff wavelength of poled polymer was about 600nm. The second order nonlinear optical coefficient

of the poled polymer was estimated as d_{33}=13.6pm/V by Maker Fringe method[11]. The quartz crystal was used as a reference sample and d_{11} of the quartz was 0.4pm/V[12]. The fundamental wavelength was 1064nm from a Q-switched Nd-YAG laser. P-polarized fundamental and second harmonic beams were used. The repetition rate of fundamental beam was 10Hz and pulse width was 10ns. The generated second harmonic wave was detected by photomultiplier after removing the fundamental beam with filter. The nonlinear optical coefficient of poled polymer had good longterm stability as shown in Fig. 2. The polymer waveguide film was formed with taper structure to satisfy the phase matching condition. The waveguide thickness for guided mode phase matching was decided by theoretical analysis using the refractive indices of poled polymer and substrate.

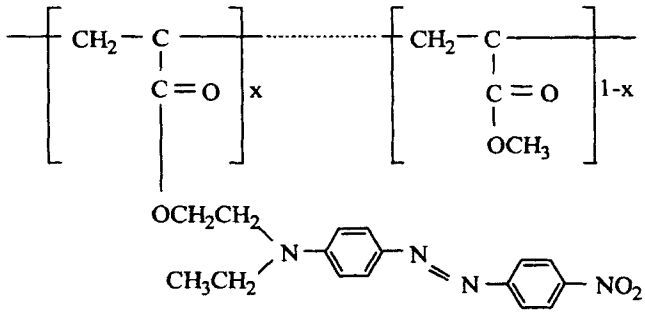

FIGURE 1 The structure of poled polymer

NEAR UV SECOND HARMONIC GENERATION

The refractive index of poled polymer observed by ellipsometer and m-line method is shown in Fig. 3. The refractive indices of poled polymer and substrate were 1.538 and 1.47 at fundamental wavelength (740nm), and 1.522 and 1.492 at second harmonic wavelength (370nm). The waveguide structure was used to generate second harmonic beam and the fundamental beam was TM polarized to use d_{33} of poled polymer. Guided mode phase matching using anomalous dispersion of poled polymer was used to generate near UV SHG according as the cutoff wavelength of polymer film used in this study was about 600nm and the poled polymer film had transparency window near UV range. The TM polarized second harmonic power $P^{2\omega}$ from the TM polarized fundamental power P^{ω} is given by

$$P^{2\omega} = \frac{2\varepsilon_0^{1/2}\mu_0^{3/2}\omega^2(N_{eff}^{\omega})^4\, d_{33}^2 S^2 L^2 (P^{\omega})^2}{N_{eff}^{2\omega} W_{eff}^{2\omega} (W_{eff}^{\omega})^2 D} \mathrm{sinc}^2\left(\frac{\Delta\beta L}{2}\right) \quad (1)$$

$$S = \int_{-\infty}^{\infty} (H_{ym}^{\omega})^2 H_{yn}^{2\omega} dx \qquad (2)$$

where m and n are mode numbers. S is the overlap integral between the normalized magnetic field of fundamental wave (H_{ym}^{ω}) and the normalized magnetic field of second harmonic wave ($H_{yn}^{2\omega}$). The propagation of fundamental beam is z-direction and the waveguide film thickness is x-direction. L is interaction length, d is nonlinear optical coefficient, and $\Delta\beta$ is phase mismatch. If phase matchig is accomplished, $\Delta\beta$=0. N_{eff}^{ω} is the effective refractive index for fundamental wave and $N_{eff}^{2\omega}$ is the effective refractive index for second harmonic wave. W_{eff}^{ω} is effective thickness of waveguide layer at fundamental wave, and $W_{eff}^{2\omega}$ is effective thickness of waveguide layer at second harmonic wave. D is beam width. The depletion of fundamental wave and the loss of poled polymer were ignored in theoretical analysis.

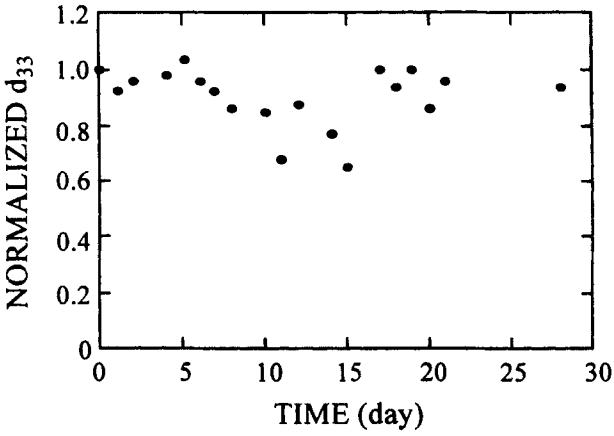

FIGURE 2 The longterm characteristics of poled polymer nonlinearity

The modal dispersion curve is shown in previous paper[10] at the fundamental wavelength (740nm) and the second harmonic wavelength (370nm). The second harmonic power calculated by using eq. (1) is shown in Table I. The nonlinear optical coefficient of the poled polymer depends on the wavelength, however, in this theoretical analysis the nonlinear optical coefficient of poled polymer at 740nm was supposed to be 13.6pm/V. The phase matching between fundamental TM_0 guided mode and second harmonic TM_0 guided mode was possible, and in this case the second harmonic power was the highest in theoretical analysis. It is the reason that the overlap integral between the fundamental TM_0 guided mode

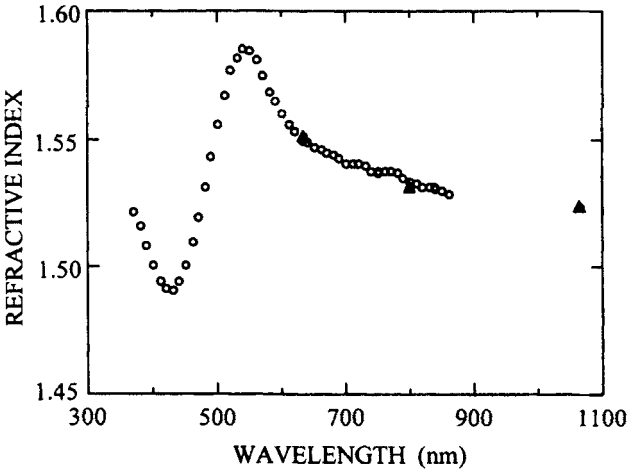

FIGURE 3 The refractive index dispersion of poled polymer. The open circle is ellipsometry data and the triangle is measurement data by m-line method

and the second harmonic TM_0 guided mode is larger than that of any other phase matchable mode between the fundamental and the second harmonic wave because both have the same mode number. In the case of fundamental TM_m guided mode and second harmonic TM_n ($n \neq 0$) guided mode, the overlap integral became small compared with fundamental TM_0 guided mode and second harmonic TM_0 guided mode because the sign of second harmonic field distribution was changed in some waveguide layer thickness. The SH experiment was carried out using the sample with tapered structure of poled polymer to satisfy phase matching. The SH power as a function of waveguide layer thickness calculated using eq. (1) is shown in Fig. 4. The SH power is oscillated around the phase matching thickness and is extremely decreased if the waveguide layer thickness is not satisfied with phase matching condition. FWHM is about 15nm from Fig. 4 when the interaction length is 1mm.

TABLE I Theoretical SH efficiency on each phase matching point

Fun. mode	*SH mode*	N_{eff}	*Film thickness (μm)*	*efficiency*
0	0	1.5156	1.124	1.05×10^{-2}
1	0	1.5209	2.971	9.13×10^{-7}
1	1	1.5162	2.594	1.65×10^{-5}
1	2	1.4974	1.80	6.3×10^{-4}

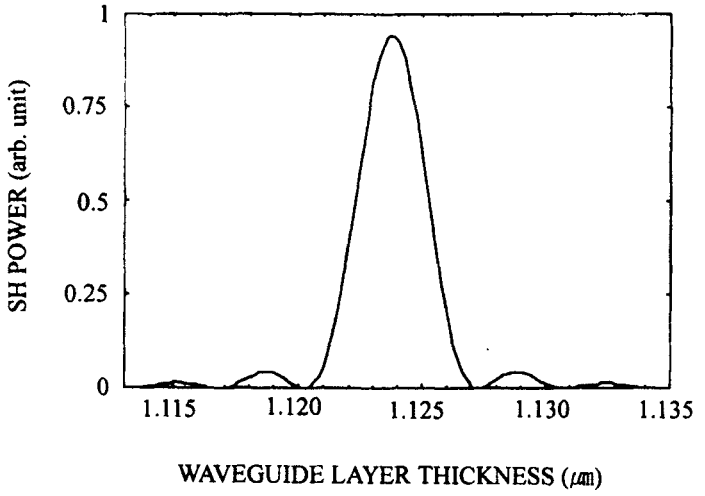

FIGURE 4 The calculated second harmonic TM_0 power from the fundamental TM_0 guided mode as a function of waveguide layer thickness

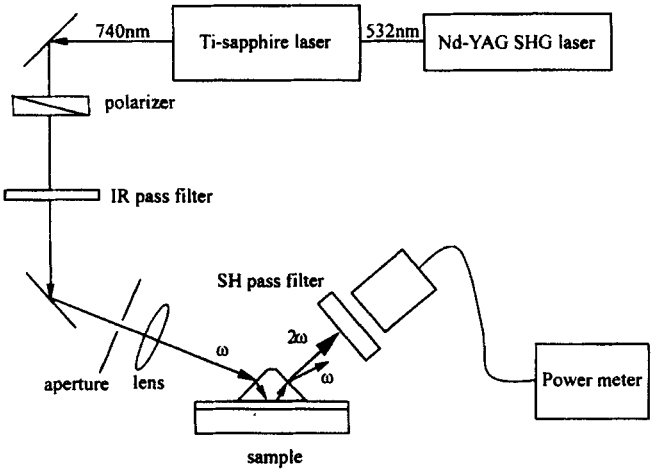

FIGURE 5 The experimental setup for SHG

The experimental setup is shown in Fig. 5. The fundamental wavelength was tuned as 740nm from Ti-sapphire laser pumped by second harmonic wave (532nm) of Nd-YAG laser (1064nm) using KD*P crystal. The sample was mounted on the X-Y and rotation stage and the fundamental beam was coupled

into waveguide layer by prism coupler through the polarizer and lens. The fundamental and second harmonic waves were TM polarized. The incident position of fundamental beam was precisely adjusted by moving the sample perpendicular to the incident fundamental beam and the SH beam was generated at the phase matching thickness as shown in Fig. 4. This thickness was satisfied with phase matching condition, i.e. $\Delta\beta = 0$, and corresponds to the intersect point between fundamental TM_0 and second harmonic TM_0 in the modal dispersion curve in previous paper[10]. The generated second harmonic wave was detected by Si-photodiode after removing the fundamental beam with filter. The measured second harmonic power on a fundamental power is shown in Fig. 6. The phase matched polymer film thickness was calculated as 1.1226μm from the prism coupling angle of the fundamental beam, which was good agreement with theoretical analysis. The second harmonic power was not very high and did not have the perfect square characteristics of the fundamental power. The reasons are considered as follows; (1)poled polymer has a little absorption in second harmonic wavelength, (2) poled polymer loss and measurement error have an effect on the SH power and (3) coupling efficiency of prism coupler is generally low.

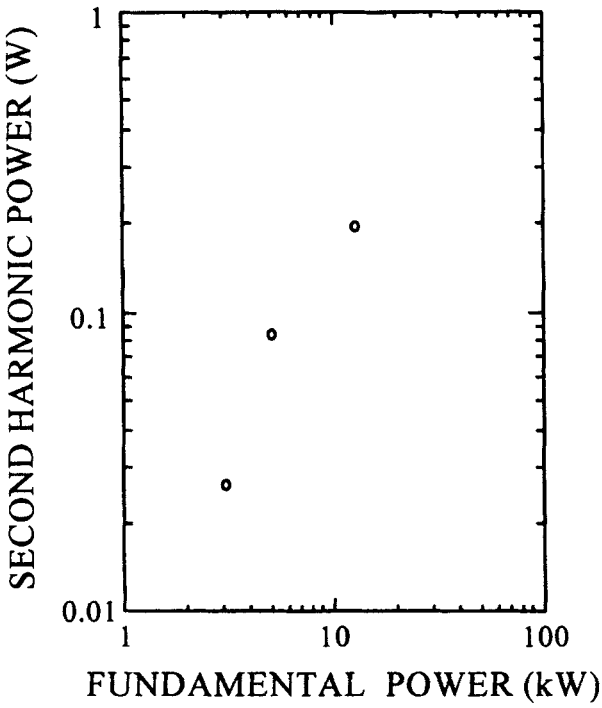

FIGURE 6 The second harmonic power as a function of fundamental power

CONCLUSION

Near UV second harmonic TM_0 guided mode (370nm) was generated from the fundamental TM_0 guided mode (740nm) in poled polymer waveguide film by guided mode phase matching using anomalous dispersion of poled polymer. The second harmonic power was the highest in the phase matchable modes. It is because the overlap integral between fundamental wave and second harmonic wave is the largest in that case. For efficient second harmonic generation, we need following conditions. The perfect transparency window in second harmonic wavelength region, easy controllability of film thickness with long interaction length, and high coupling efficiency of fundamental beam into waveguide are required. And finally low loss of materials, which is one of the most important problems in especially polymer, is required.

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